

COMPARISON OF REPROCESSING METHODS FOR LIGHT WATER REACTOR FUEL

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Sharon (Jess) Chandler

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COMPARISON OF REPROCESSING METHODS FOR LIGHT WATER REACTOR FUEL

Approved by:

Dr. Nolan Hertel, Committee Chair
School of Nuclear and Radiological
Engineering
Georgia Institute of Technology

Dr. Daniel Tedder
Department of Chemical and Biomedical
Engineering
Georgia Institute of Technology

Dr. Ratib Karam
School of Nuclear and Radiological
Engineering
Georgia Institute of Technology

Date Approved: 20 November 2006

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SUMMARY

Currently, the United States is investigating methods to close the nuclear fuel cycle and increase the use of nuclear power for electricity and cogeneration applications. Congress has called for and held hearings in an attempt to determine an appropriate path forward for reprocessing of nuclear fuel. However, each current proposed method presents a different set of attributes with regards to: Complexity, safety, wastes, and proliferation risks.

This thesis provides a decision analysis methodology for approaching the reprocessing issue. The presented methodology builds on the previous work done in the 1970's. Further, current reprocessing technologies which are capable of processing the oxide fuels utilized in the majority of United States reactors are evaluated across ten attributes related to reprocessing performance. A weighted total score is provided for each reprocessing method in order to separate the technological from political or emotional issues related to selection of a process.

While it is not the goal of this thesis to select a particular 'best' technology, application of this methodology results in the selection of the COmbined EXtraction (COEX) technology when equal weighting is put on the attributes as defined. It is unlikely that a decision maker will approach this decision with equal weighting; however, actual decision maker weightings are known only to the decision makers. By altering the weightings, different technologies are selected.

CHAPTER I

INTRODUCTION

Reprocessing of used nuclear fuel will be necessary to continue nuclear power into the future. There are a multitude of possible methods to employ in a future fuel cycle. Several instances in the literature focus on the operation of a future fuel cycle, be it a combination of fast and thermal reactors, a fully fast reactor fuel cycle, a continuation of open fuel cycle practices, or another idea, like the DUPIC process where used light water reactor (LWR) fuel would be directly input to heavy water reactors [30, 57, 77, 49]. A necessary component of all but the open fuel cycle continuum require a reprocessing technology which can satisfy technical and political requirements, and some argue that the open fuel cycle option would benefit by separations as well [39, 74]. The sheer volume of spent nuclear fuel stored and produced in the United States is cause for interest in separation of usable materials; based on the reported legacy stored fuel and a rate of 2,000 metric tons of heavy metal (MTHM) used commercial fuel per year, there will be over 55,000 MTHM used nuclear fuel in the U.S. by the end of this year [14].

In this paper, reprocessing methods which can separate components of used nuclear fuel are compared over several attributes. This comparison is accomplished through Multiattribute Utility Theory which is a common tool for decision analysis.

1.1 Why a decision methodology?

A decision methodology is necessary for consistency, transparency, and clarity. Due to the special significance of nuclear fuel, ultimate decisions for a particular reprocessing technology in the United States will be made in the public realm. This does not mean that citizens will vote for a technology, but more that the path forward will ultimately be chosen by elected or appointed government officials rather than industry. In fact, the Energy and Water Development Appropriations Act for fiscal year 2006 includes a House report (109-086) requiring the Department of Energy to recommend a specific reprocessing technology by

2007 [23].

As with any technically intensive policy decision, there is a problem for lawmakers when the technical experts are not in agreement, “How can politicians tell the public which experts to believe?” [12]. Also, how are lawmakers to find the time to understand the arguments given by conflicting scientific or technical experts advocating different sides, “political leaders will not be able to spend the time necessary to understand scientific debates in sufficient depth to distinguish the relative validity of positions taken by sophisticated advocates” [52]. LaPorte (1978) argued that lack of information and attention to the consequences of radioactive waste management forces speculation in public debate, and he stated, “careful description of the technical alternatives is necessary to proceed to the next step” [65].

There also exists the problem that technical advisers to political decision makers often become involved in the political as well as moral issues related to the scientific or technical evaluation at hand. “[W]e must recognize that the moral responsibility which many scientists feel very deeply can easily affect their judgement as to the state of scientific fact when the scientific facts are not yet crystal clear . . . do not believe it is possible for scientists to have deeply held moral and political views about a question and simultaneously maintain complete objectivity concerning its scientific components” [52]. Morality must be considered; however, an attempt should be made to separate the technical and moral, political, or emotional issues. The presented methodology attempts to do address this issue.

Further, Lagus (2005) acknowledges, “One of the mistakes made during early nuclear reprocessing was the relative immaturity of the technology at the time of deployment. The U.S. reprocessing industry has become nonexistent because of this mistake, and therefore the country must remain cautious to prevent similar errors that could damage the reputation of the technology. Deployment should not take place until a fully realized solution can be found” [62]. While questions surround ‘what might have worked’ regarding specific causality of the decline in reprocessing in the U.S., Lagus’ point is well made for the surrounding uncertainty and need for a firm solution.

1.2 Overview of analysis

This analysis uses several overarching issues, related to reprocessing of used nuclear fuel, to compare the ten different technologies shown in Table 1. Each issue is broken down into the lowest level technical attributes. The low-level attributes are combined to form an overall score for each technology. The premise behind such an organization of technical points is to break the technical components (attributes) away from the political (and social or emotional) components (weights).

Table 1: Technologies Evaluated	
Aqueous Processes	Pyroprocesses
PUREX	LiCl-KCl
COEX	NaCl-KCl
UREX	Flouride Volatility
UREX+1	FLUOREX ^a
UREX+2	
Supercritical CO ₂	

The political components can be added by assigning weights to various attributes when developing the overall scores. Part of multiattribute utility theory is quantifying the decision makers' preferences with regards to importance of attributes [47]. This analysis offers equally weighted attributes as a non-influenced nominal case. Also, some manipulation of weightings is offered for example and discussion.

1.3 Intended use of analysis

This analysis is intended to be a dynamic approach which can be used by decision makers when approaching the problem of selecting a specific reprocessing technology. Decision makers can modify the weightings to select a technology which meets all of their required parameters. Chairwoman Biggert addressed a hearing before the House committee on science, subcommittee on energy on this issue, "Technology decisions for reprocessing must take into account technology and policy decisions for the entire fuel cycle" [24].

It is beyond the scope of this report to analyze the multitude of possibilities which exist when considering the entire fuel cycle; however, it is expected that decision makers can assign utility to reprocessing issues based on the effects or needs in the rest of the fuel

cycle. Because each decision maker may have different utility rankings (i.e. one may see proliferation risk as the most important determinant of technology while another may see flexibility as the most important determinant), the analysis allows for different assignment of weight, or utility, to each attribute. For example, when Shane Johnson, Department of Energy Office of Nuclear Energy, was asked by the subcommittee hearing, “How will DOE select a reprocessing technology for the future? What factors will be taken into account?”, he replied (in a written statement), “The selection of a reprocessing technology is dependent on economics, reliability, ease of scale-up and considerations related to safety and proliferation resistance. Advanced aqueous processing are the best suited to treat spent nuclear fuel being stored and generated today...Pyrochemical processes may be better suited for spent fuel from advanced fast reactors” [24]. No clear preference was stated, but inferred utility could select that the weight is higher for economics than for proliferation resistance based on order of statement; the author is doubtful that this is the case. Because true preferences are not always directly stated, it is important to have flexibility in the analysis method.

Due to the previously mentioned Energy and Water Development Act requirements, the ultimate decision on reprocessing technology in the United States will be made in the executive arena, at the Department of Energy. The decision makers within the executive branch have been described by Hamlett (1991) as “largely insulated from changing political trends, [but] their agencies are often caught up in presidential politics and the alterations in party control of the administration.” [42]. The 2007 deadline is still within the timeframe of the current administration, so a political change is not expected to influence this decision; however, a new administration may review the decision or negate it altogether. It is not clear how the preferences of the decision makers in the executive compare to those in the legislative or the public arenas.

CHAPTER II

BACKGROUND

This thesis attempts to provide a decision method for reprocessing technology. While there has been quite a bit of recent technical work on reprocessing methods for used nuclear fuel, the current literature does not address how a process will be chosen for implementation. In fact, most papers on reprocessing methods discuss only the experiment at hand and do not put their work into the context of a nuclear future.

An overview of reprocessing is presented to bring this paper in context. A methodology for comparing reprocessing methods which was originally published in 1978 by Selvaduray and Goldstein, and not addressed significantly since then, provides a foundation for the presented methodology, and the former is described in this section. The science of decision analysis provides theoretical standing for investigating a framework for such a methodology; a brief review of part of this theory is provided at the end of this chapter.

2.1 Overview of Reprocessing

Reprocessing of spent nuclear fuel refers to a separations process with the function of fractionation. Separation processes can be broadly described as “any set of operations that separate solutions of two or more components into two or more products that differ in composition”, and separation is “achieved by exploiting chemical and physical property differences between the substances through the use of a separating agent” [3]. While some of the technologies have changed over the years, the main technical objectives for conducting these separations are still the same as stated by Benedict (1981), “1)recover uranium and plutonium, and thorium if present, for reuse as nuclear fuels; 2) to remove radioactive and neutron-absorbing fission products from them, and 3) to convert the radioactive constituents of spent fuel into forms suitable for safe, long-term storage.” [10]. The motivation for the continued processing of used nuclear fuel can be found in Table 2 as the bulk of used nuclear fuel is reusable material.

Table 2: Composition of Used Nuclear Fuel

Element or group	Percent, by weight
uranium	95.6
fission products, stable and short-lived	0.3
plutonium	0.9
cesium and strontium	0.3
minor actinides (Np, Cm, Am)	0.1
iodine and technetium	0.1

2.1.1 Reprocessing history

Originally, reprocessing was carried out with a bismuth phosphate process developed at the Oak Ridge National Laboratory and utilized at the Hanford, Washington site beginning in 1944 [31]. This method required several stages including dissolution of the cladding material and precipitate formation along with centrifuge steps. The dissolution step removed the cladding from the fuel. The fuel material, in nitric acid, would then be treated with sodium nitrate for reduction of plutonium; $BiPO_4$ would precipitate tetravalent plutonium. Uranium and most of the fission products were discarded as a heavy metal waste. The plutonium was further purified in decontamination steps. Disadvantages included not separating uranium, operating in batches, and large amounts of waste and chemicals [10].

A new process was developed at Argonne National Laboratory (ANL) which used REDuction and OXidation (REDOX). This aqueous process involved aluminum nitrate in the aqueous phase with hexone as the solvent for extraction. It led to less waste, more complete extraction, and continuous operations which were seen as advantages over the bismuth phosphate process, but had a drawback of large amounts of aluminum nitrate being used [13] REDOX, or a modified REDOX process, was used at Hanford and at Idaho Chemical Processing Plant. The disadvantages “were the volatility and flammability of the hexone solvent and the large amounts of nonvolatile reagents such as $Al(NO_3)_3$ added to the radioactive wastes” [10].

Scientists at Oak Ridge National Laboratory also developed the plutonium and uranium extraction (PUREX) process which is used throughout the world today. All commercial reprocessing plants in operation today use this technology. PUREX reprocessing involves a

bath of nitric acid and solvent extraction of plutonium and uranium with tributyl phosphate (TBP) in a hydrocarbon diluent [10]. Adaptations have been applied to the PUREX process to make it more useful for treatment of waste and civilian use. With the addition of a second extraction stage, minor actinides can be removed for waste treatment. Modifying the process in 1999, U.S. scientists extracted only uranium, making the process called uranium extraction (UREX). The UREX process adds acetohydroxamic acid (AHA) which reduces the extractability of plutonium and neptunium [53].

Initial research in pyroprocessing was carried out by the Department of Energy for treatment of spent fuel from Experimental Breeder Reactor II (EBR-II). A melt-refining process was developed which recovered actinides while removing volatile fission products from this metal fuel [102]. Because this process provided only offered partial separation of actinides and was not able to separate plutonium which was considered desirable at the time, it was not adopted [63].

2.1.2 Reprocessing today

Reprocessing of nuclear fuel is a globally existing technology, regardless of the specific process utilized. By 2005, more than 75,000 MTHM spent nuclear fuel had been processed world-wide [79]. Broadly, the technologies for separating/fractionating used nuclear fuel fall into either aqueous or non-aqueous processes, as shown in Table 1 (Introduction). Each of the examined technologies in this analysis are described in detail in the Technologies chapter.

While it is generally expected that reprocessing of used nuclear fuel will be followed by additional use of the fissile and/or the fertile nuclides, there is not a consensus as to which direction is most likely. It is beyond the scope of this analysis to evaluate the feasibility of a particular advanced fuel cycle or reprocessing of fuel which differs from the current oxide fuels. The nuclear community does express that reprocessing of additional cycles will present new technical challenges; for example, considering Mixed Oxide Fuel (MOX): “Reprocessing of a spent MOX fuel load is technically feasible but much more difficult than reprocessing of conventional UO_2 spent fuel . . . increased criticality risk during dissolution,

increased solvent degradation due to higher alpha emissions, DBP complexes strongly with Pu species which increases complexity, heat generation increases, and leads to need for remote fuel fabrication” [6].

2.2 Detail on previous methodology

The previous comparison method presented by Selvaduray and Goldstein (1978), is heavily drawn from in order to present a summary in this section [37, 97, 96]. Their comparison pooled eleven weighted parameters into an overall ‘performance factor’ for fourteen reprocessing technologies, allowing for comparison of methods, see Table 3. The current study will build on this work by updating the method and providing for a more stable approach by rooting in decision analysis theory.

These parameter scores were leveled to a range of 0-10 and weighted with a rate (r) and with a level of importance (R) that was determined by surveying ‘knowledgeable persons’ in the field to arrive at an overall score, or ‘performance factor’ (E) for each of the fourteen technologies considered; the performance factor is a ratio of the ranked ratings of a technology to the maximum possible. Surveys were distributed amongst several different groups to analyze the relative importance of separate parameters to different groups.

$$E = \frac{\sum rR}{10 \sum R} \quad (1)$$

This previous analysis went on to speculate as to future fuel use and reactor operations with an attempt to determine the best reprocessing technology for the year 2000 under different futures. This comparison provided an important contribution to the field on two accounts: Conceptualization of the important aspects of decision making in reprocessing and compilation of related factors for the technologies of the day. However, the comparison lacks internal validity as some of the inputs are repeated in separate sections with different weights, and it lacks flexibility for decision makers to assign their own utility functions to each of the lowest level attributes. Hopefully, the presented methodology will expand the ideas of the previous authors correctly without repeating these short-falls.

Table 3: Parameters and Definitions in Previous Method

Parameters	Defined by
Technical Complexity	$\frac{\text{NUMBER OF UNITS IN MOST COMPACT PROCESS}}{\text{NUMBER OF UNIT OPERATIONS IN PROCESS CONSIDERED}} \times 7$ + additional points for remote refabrication
Safeguards of Strategic Nuclear Materials	Rating composed of matrix depending on decontamination factor, labor intensity, and effluent streams
Waste Volume	Rating assigned based on form and relative quantity of effluent wastes
Sensitivity to Changing Regulations	Determined based on the ratings assigned to the technology for risk to population, safeguards of SNM, and waste volume
Sensitivity to Change in Fuel Type	Rated between 3 and 10 depending on number of fuel types handled
Maintenance	$\frac{\text{CAPITAL COST}}{\text{NORMALIZING CONSTANT}} + \text{QUALITY FACTOR}$
Stage of Development	Scaled from 1 to 10, conceptual to commercial
Reliability	based on factors of heat, radiation damage, and principle of separation
Risk to Population	Rating based on ‘total toxicity’ between 10^9 and 10^{10}
Economic Advantage	Rating from 1, highest cost, to 10, lowest cost, based on author’s calculations
Decontamination Factor	Rating from 1 (DF of 10^2) to 10 (DF greater than 10^6)

2.3 *Decison Analysis*

“A decision problem is one with more than one available course of action... [the decision maker] must choose a ‘best’ course of action from the alternatives ... consequences can rarely be described completely in terms of one attribute” [54]. Determination of a reprocessing technology to follow for the near future in the United States is fundamentally a decision problem with multiple attributes and multidimensional consequences associated with any choice. The current method is based on Multiattribute Utility Theory (MAU). Both the science of decision analysis and MAU will be described here.

2.3.1 Science of decision analysis

Decision analysis consists of, “structuring the problem (which includes specifying objectives, building a value hierarchy and establishing attributes for the lowest-level objectives); identifying the feasible alternatives, their impact and uncertainty (if necessary); quantifying preferences (which includes the assessment of component attribute utilities as well as the value trade-offs); evaluating alternatives and performing sensitivity analysis” [47]. Often, decision scientists are looking back on a decision which was made to understand the reasoning behind a choice; in this case, the goal is to look forward to structure transparency into the decision as it is made.

The importance of decision science in situations of dynamic increasing returns is best demonstrated by investigating the outcome following the lack of comprehensive analysis before moving forward with light water reactors. The events leading up to the advent of civil nuclear power operations “caused a civilian power project to be rushed forward, before the physicists involved were ready to make a choice among the available technologies, effectively forcing the choice of light water” [25].

2.3.2 Multiattribute Utility Theory (MAU)

Multiattribute Utility Theory allows for decisions to be made on parameters without conversion to an economic cost basis. MAU is most desired when “good cost data are not available or when cost is not suitable as a measure of performance” [17]. For some of the

comparisons related to reprocessing technology, cost certainly is not a suitable performance measure, and attempts to associate cost with the comparable issues would induce more error. Application of cost for something that may occur at some unknown point in the future also could incur errors in the discount rate which may lead to not pursuing a good choice. “[I]t is fallacious to make long-term decisions on the basis of current costs unless these costs are intrinsic. It would be a little like abandoning the development of jet aircraft on the basis of 1939 costs” [101].

The general form of the multi linear utility function is the beginning point for application of MAU, but the function will be simplified down to the additive model for this analysis. The proofs behind these simplifications are published in some detail [17, 55, 54], and will not be presented here. The general form, when $X = (x_1, x_2, \dots, x_n)$ is a vector of random variables over performance measures; i, j , and m are separate attributes; u_i is a single attribute utility function (scaled from 0 to 1); ω_i is the weight over $0 \leq \omega_i \leq 1$, for all i ; and ω_{ij} and ω_{ijm} are constants of interaction between parameters, the Multattribute Utility $U(X)$ appears as equation 2.

$$\begin{aligned}
 U(X) = & \sum_{i=1}^n \omega_i u_i(x_i) + \sum_{i=1}^n \sum_{j>i}^n \omega_{ij} u_i(x_i) u_j(x_j) + \\
 & \sum_{i=1}^n \sum_{j>i}^n \sum_{m>j>i}^n \omega_{ijm} u_i(x_i) u_j(x_j) u_m(x_m) + \dots + \omega_{123\dots n} u_1(x_1) u_2(x_2) \dots u_n(x_n)
 \end{aligned} \tag{2}$$

Understand that Equation 2 refers to overall utility of a decision which would entail a combined score for all the choices within the decision. Technologies ‘ i ’, ‘ j ’, and ‘ m ’, etc. are included in this equation as well as the decision maker’s utility of each ω and the interactions between them.

Utility independence allows for simplification of the general form. The initial theorem and proof for the simplification was provided by Keeney [55] and is summarized here. Let $U(X) = u(x_1, x_2, \dots, x_n)$, where U is a utility function and x_i is an attribute of that function. If vectors Y and Z are defined such that $X = Y \times Z$, and the decision maker has the same “relative preference over lotteries on Y , when Z is held fixed at z_0 ” for any value of z_0 then Y is *utility independent* of Z . This implies that the impacts are equal, but not

necessarily zero. The general form under utility independence reduces to the multiplicative model (equation 3).

$$\begin{aligned}
U(X) = & \sum_{i=1}^n \omega_i u_i(x_i) + \sum_{i=1}^n \sum_{j>i}^n \omega \omega_i \omega_j u_i(x_i) u_j(x_j) + \\
& \sum_{i=1}^n \sum_{j>i}^n \sum_{m>j>i}^n \omega^2 \omega_i \omega_j \omega_m u_i(x_i) u_j(x_j) \times u_m(x_m) + \dots + \omega^{n-1} \prod_{i=1}^n \omega_i u_i(x_1)
\end{aligned} \tag{3}$$

Additive independence allows for further simplification of the model, given that the interaction terms, meeting the condition of equality under utility independence, are equally zero. If the interaction terms are zero, it is implied that there is no preferential interaction. Previous studies have shown that the additive MAU model can be used even when the preferential interaction is unknown, as in this case, and will “typically provide good approximation of preferences that do not satisfy additive independence” [17]. The additive form shown in equation 4 is simplified down to utility for one parameter, technology in this case, because interaction has been removed.

$$U(X) = \sum_{i=1}^n \omega_i u_i(x_i) \tag{4}$$

Note: The summation of the weights, ‘ ω_i ’, for all i must equal 1.

The importance of defining the decision maker’s utility has been stressed in the literature. Clearly, the utility will depend on who is defined as the decision maker. For the purposes of this analysis, the decision maker is constructed to be any individual or group who might want to apply their preferences to this model to determine the ‘best’ reprocessing technology. Because of the separation of technical and political or emotional issues, selection of a reprocessing technology is possible without great depth of knowledge of the chemical or nuclear engineering problems involved. With such a broad goal, it is impossible to apply a particular utility to the model in this paper and assume that the selected technology is certainly the one which should be pursued. Instead, the results related to variations of utility preferences are considered in the Discussion chapter.

2.4 Limitations of this method

While the goal of this thesis is to provide an objective decision method for choosing a reprocessing technology, the method is still subject to author bias due to the attributes chosen for inclusion. There are many possible attributes that could be included, but some have been ignored for simplicity. Additionally, numbers used to determine attribute values in the present methodology are in some cases approximated or average values - actual values for a particular plant or batch could differ from the values used. This method is further limited by the temporal differences in reprocessing technique experimentation; processes presented in the past may experience bias due to technological advances which would have been incorporated in the process if the process were developed today. Limitations should be considered when applying the presented decision methodology to reprocessing choice.

CHAPTER III

METHODOLOGY

The present method modifies the comparison method introduced in 1978 to make the comparison more dynamic and applicable today. The major changes involve removal of some attributes which are repetitive or non-quantifiable, application to a different set of technologies, and introduction of multiattribute utility theory. Decision science offers that “A complete model would present (a) a measure of the efficiency of the A_i at attaining the goals, (b) some measure of the weight of the goals when the goals are in conflict, (c) a criterion of adequacy for the operation O for choosing A_j ” [9]. Each of these are addressed in this methodology.

3.1 Issues

Several overarching issues are addressed, and their lowest-level technical attributes are considered and scored (to allow for equivalent ranges) for each technology. A final score is determined by performing a weighted summation of the scores. Two of the overarching issues (Economics and Stage of Development) which are addressed are not scored since they are a function of the other issues or vice-versa. Each attribute is levelized to a value between 0 and 1 to facilitate comparison, with 1 being the best conceivable for each attribute. Technological advances could require reconsideration of the ‘best conceivable’ value which could affect different scores for the technologies under consideration.

3.1.1 Technical complexity

The degree of technical complexity in a process is necessarily considered in a comparison of technologies. Studies which attempt to estimate costs before a complete design is prepared often consider these factors. All processes involving irradiated nuclear fuel are technically complex, but there are aspects which effect the degree of complexity; for the purposes of comparison, the attributes considered are: Number of steps in the process, operating

temperatures, operating pressures, and use of corrosive chemicals. The author notes that other conditions may additionally need to be considered in this fashion in future work, including other energy inputs or requirements for special atmospheric chemistry (i.e. argon atmosphere).

3.1.1.1 Number of steps

A process which achieves the desired outcome in fewer steps provides several advantages over a greater-step process. These advantages can include: decreased risk of failure, ease of duplication, and reduced costs. The steps counted do not include cladding removal or chop processes or any treatment following the main separation process, except for UREX+1, UREX+2, and FLUOREX which are defined by their additional steps. To be specific, the first step counted is the one following fuel chopping, and the last step counted is decontamination of the selected element(s); parallel separations are counted separately (i.e. decontamination of U and decontamination of Pu would be two steps). Additionally, multiple stages within a solvent extraction process are not counted; solvent extraction steps are counted in broader terms of extraction, scrub, and strip. The ‘best’ possible number of steps would be one step. The score for number of steps, ‘NS’, for each technology, ‘j’, can be defined by equation 5.

$$NS_j = \frac{1}{n_j} \quad (5)$$

3.1.1.2 Operating Temperatures

Recognizable differences in complexity exist dependent upon the degree to which the process conditions must deviate from ambient conditions [105]. Processes are evaluated against ambient temperature conditions. Due to obvious reasons for non-standard ambient conditions, the basis for ambient is assumed to be 100 degrees celsius, or 373.15 K, at standard pressure. This particular definition of ambient temperature could be argued (i.e. Why not use room temperature?); standard temperature is defined by National Institute of Standards and Technology (NIST) at 273.15 K, or 0 [81]. With 373.15 K as ambient, heating and cooling appear on opposite sides of ambient and can be compared for their deviation

alone. The attribute for operating temperature, ‘OT’, for each technology, ‘j’, can be defined by 1, if the technology is known to operate at ambient temperature, and by equation 6 if temperature must reach or be held at some defined value.

$$\begin{aligned} OT_j &= \frac{373.15K}{T_jK} \forall T > 373.15 \text{ K} \\ OT_j &= \frac{T_jK}{373.15K} \forall T < 373.15 \text{ K} \end{aligned} \quad (6)$$

Note: The temperatures used will be minimum or maximum values that must be attained.

3.1.1.3 Operating Pressures

Much like the discussion for temperature, above, deviation from ambient pressure, either to extremely high pressures or to vacuum conditions, leads to increased complexity. Thus, the calculation will consider the deviation from standard pressure at 1 bar, 750 torr. The attribute for operating pressure, ‘OP’, for each technology, ‘j’, can be defined by 1, if the technology is known to operate at ambient pressure, and by equation 7 if the pressure must reach or be held at some defined value.

$$\begin{aligned} OP_j &= \frac{750\text{torr}}{P_j\text{torr}} \forall P > 750 \text{ TORR} \\ OP_j &= \frac{P_j\text{torr}}{750\text{torr}} \forall P < 750 \text{ TORR} \end{aligned} \quad (7)$$

Note: The pressures used will be minimum or maximum values that must be attained.

3.1.1.4 Use of corrosive materials

A process which has corrosive products as part of its operation will be more likely to be in need to preventative or corrective maintenance. Maintenance poses a particular problem for facilities which are processing irradiated nuclear fuel due to shielding requirements, risk to laborers, and additional organizational burdens to operate over maintenance periods. For this attribute, the scaling method used in the ‘quality factor’ for the previous comparison’s maintenance rating is extended [96]. However, the rating for corrosion, ‘CM’, is changed from a 0 to 3 scale to a 0 to 1 scale for consistency. Any technology with no corrosive

agents will be scored unity, as this is the best possible scenario. As shown in equation 8, a technology with high corrosion due to the materials involved will be scored 0.1, and corrosion from nitric acid will score 0.5. The lack of available data and the intense research in the materials sciences both contribute uncertainty to the degree to which corrosion will continue to exist in a certain process.

$$\begin{aligned}
CM_j &= 0.1, \forall \text{HIGH CORROSION} \\
CM_j &= 0.5, \forall \text{NITRIC ACID CORROSION} \\
CM_j &= 1 \forall \text{NO CORROSIVE AGENTS}
\end{aligned} \tag{8}$$

3.1.2 Safety

Safety is a very broad consideration; for this analysis, safety is defined as meaning the general safety of the technology to the population. There are many scenarios which could be drawn up (and sometimes have been) in regards to safety of a reprocessing plant; however, most scenarios, like the possibility of a high-speed aircraft collision to a sensitive plant section, are conceivably applicable to all reprocessing facilities equally. Risk analysis calculations have suggested that other non-nuclear facilities pose a greater risk in such events, Liquid Natural Gas facilities, for instance [49]. Safety attributes are based on the maximum credible accident (MCA) and the likelihood of explosion.

3.1.2.1 *Maximum credible accident*

For some of these processes, accident studies have not yet been conducted; however, the nature of the process allows for determination of a characterization of the possible materials included in an accident. The attribute ascribed is based on the maximum total toxicity related to the MCA; the risk of the MCA is expected to be similarly low for all processes due to imposed standards. Therefore, the danger that is considered for the general population is based on one MCA occurring with the same probability for all technologies.

The previous method addressed total toxicity as a function of the gaseous, liquid, and solid toxicity [96], distributing the score over the range of total toxicities (all compared technologies had a total toxicity on the order of 10^9). This present analysis will utilize

the total toxicities presented in the former comparison because they are valid measures of possible release under MCA which the previous author already formulated into relative toxicities; however, the scoring for ‘MCA’, equation 10, is scaled to be from 0 to 1 for consistency. The scaling is such that a toxicity of 1×10^{10} would score zero and 1×10^9 would score one.

$$\begin{aligned} \text{TOTAL TOXICITY} = & \text{RELATIVE GASEOUS TOXICITY} + \\ & \text{RELATIVE LIQUID TOXICITY} + \\ & \text{RELATIVE SOLID TOXICITY} \end{aligned} \quad (9)$$

$$\begin{aligned} \text{FOR TOTAL TOXICITY} &= P \times 10^9 \\ MCA_j &= \frac{1}{P} \end{aligned} \quad (10)$$

3.1.2.2 Explosiveness

There is also the possibility of explosion for some materials which are utilized in reprocessing. For this reason, the presence of explosive materials will be an additional attribute. The explosion risk included here is not that of possible criticality explosion; engineering methods exist for preventing criticality, including geometric and chemical considerations. Rather the consideration is of an accidental chemical explosion somewhere within the process. This attribute, ‘EX’, equation 11, is ascribed a true or false score of 0 or 1 for the employment of explosive materials.

$$\begin{aligned} EX_j &= 0, \forall \text{CASES WITH EXPLOSIVE} = \text{TRUE} \\ EX_j &= 1, \forall \text{CASES WITH EXPLOSIVE} = \text{FALSE} \end{aligned} \quad (11)$$

The likelihood of explosion presented here is dealing only with the presence of explosive chemicals. There exists the possibility of heat related ‘explosive release’ of radioactivity due to dry out of high active waste tanks. While these tanks have been shown to be more vulnerable than other areas of reprocessing for release in the event of loss of cooling, engineers have determined that it is inconceivable, without sabotage, that some way of

restoring cooling could not be accomplished in the time it would take for the release to be imminent (around 5 to 6 days) [49].

3.1.3 Secondary Waste

The bulk of the arguments for reprocessing address either resource management or waste minimization directly. Thus, it would be contrary to the purpose, excepting the condition where resource reuse is the only purpose for reprocessing, trumping other considerations, to generate additional amounts of waste.

The attribute for secondary waste generation assumes that each process will necessarily emit the same amount of gaseous waste because the gaseous fission products are present in the same quantities and are not shown to undergo phase change to remain in liquid or solids in these processes. The previous method [96] employed scaling which arbitrarily positioned the AIROX process as a 10 and the REDOX process as a 1 with other processes scaling from 2-5 if they produced liquid wastes and 6-9 if they did not produce liquid wastes.

The current method still penalizes liquid wastes, but does so in a different manner. First, the total volume of waste, liquid and solid, per kgHM processed is summed and ranked from 0 to 1; then, the percent of the total volume that is liquid is calculated (percent will be between 0 and 1). The attribute for secondary waste, ‘SW’, is the average of these two numbers as shown in equation 12. When summing the volumes, all liquid wastes are assumed to have the density of water; although all liquids do not have the same density as water, it is expected that the composition of liquid wastes for each technology will be similar enough to not induce great errors into the score.

$$SW_j = \frac{\left(1 - \left(\frac{VOL_j}{VOL_{max}}\right)\right) + \left(1 - \left(\frac{VOL_{jliq}}{VOL_j}\right)\right)}{2} \quad (12)$$

3.1.4 Proliferation risks

Proliferation is an overarching concern which has found its way into mainstream conversations. Clearly, there is a concern that nuclear materials will become weapon fodder for those who would do us harm. Unfortunately, the ideas surrounding proliferation have changed since the first discussions of this term. Proliferation concerns used to involve sovereign

rogue nations which openly or clandestinely developed nuclear weapons. For the most part, national programs can be monitored with satellites and intelligence due to the extensive chemical processing required to develop weapons grade materials; evidence to this is provided by the recent chain of actions toward Iran and North Korea.

Now, proliferation concerns are more focused on the threat of sub(or multi)-national terrorist groups. The most likely method of obtaining nuclear materials for these groups is through takeover of materials during transportation or through siphoning materials from a facility. Thus, it could be postulated that locally operated reprocessing would be preferable, from proliferation standpoint, to one which requires transport.

However, the risk of a hostile taking are not isolated to one technology or another, and security is likely to be at the same level regardless of technology; additionally, siting is not technology specific. Due to these leveling characteristics of the majority of the proliferation concerns, the risk is only evaluated on the ease of weapons creation from the output streams of a technology over the input stream of used nuclear fuel (SNF). In order to calculate the proliferation risk for nuclear weapons, 'P', presented by each technology, the following ratio could be used:

$$P_j = \frac{CriticalMass_j(kg)}{CriticalMass_{SNF}(kg)}$$

Additionally, the author proposed a method of proliferation risk based on a calculation of energy intensity similar to that of the separative work unit (SWU) which is used for enrichment calculations. Discussions with colleagues suggested that this method is not practicable because not only are specific weapons structure assumed, but a prescription for development of such a weapon is inherently included. Although the author does not have detailed descriptions of weapons design and development (nor are they available to the public [74]) for which to provide a prescription, the argument against such a method is understandable.

Regardless, the use of traditional analysis of proliferation resistance is not applicable when 'dirty bombs' are taken into consideration. 'Dirty bombs' are a combination of conventional explosives with some radioactive material. While these devices are NOT nuclear weapons, and they do not possess the extreme destructive capability of nuclear weapons,

they are still dangerous [82]. These weapons also present more complexity for detection and avoidance measures.

The previous comparison [96] included four parameters, in addition to separation of a pure plutonium stream, into the evaluation of proliferation risk: Location (on or off-site), Decontamination Factor (high, medium, or low), Labor Intensity (high or low), and Effluent Streams (several or few); those methods which had pure plutonium streams were scored between 1 and 5 while those without an independent plutonium stream were scored between 6 and 10. As previously stated, I argue that any of the technologies could be operated on or off-site; siting is a political as well as an economic decision. Also, any technology could be made to be labor intensive; labor intensity could also be a function of number of effluent streams. As such, the three remaining factors which should be considered for proliferation risk are: separation of a plutonium stream, decontamination factor, and number of effluent streams.

3.1.4.1 Separation of plutonium stream

The attribute for separation of a plutonium stream, ‘SP’, is true or false logic, as shown in equation 14.

$$SP_j = 1, \forall \text{PLUTONIUM STREAM}=\text{FALSE} \quad (13)$$

$$SP_j = 0, \forall \text{PLUTONIUM STREAM}=\text{TRUE}$$

The idea that separation of plutonium results in a zero for this score should not incite those who believe separating plutonium is desirable. In fact, such persons could state such preferences and define SP'_j as $1-SP_j$ which would result in opposite values.

3.1.4.2 Decontamination factor

The decontamination factor (DF) is the degree of purification, defined as “ratio of a stated impurity to desired component ... divided by the equivalent ratio in the product” [29]. The desirability of a low or high decontamination factor can depend on the expected use of the ‘purified’ material.

For the purposes of proliferation resistance, specifically for diversion of material, fission products which ‘contaminate’ a material by making it impure can be considered a good thing. Fission product contamination can increase the dose rate of the material and also provide detectable energy signatures. However, the protection offered by these contaminants may not make the material hot enough to deter diversion attempts, as calculations show that plutonium is not significantly self-protected by fission product impurities, including cerium and europium, after the first decade following discharge [51]. Self-protection requirements put forth by the International Atomic Energy Agency are 1 Sv/hr; the “mean lethal dose”, causing death in 50 percent of a population is 3-4 Grays (an unweighted measure of radiation dose) [41]. So, the self-protection requirements expect for an unshielded contact period by diverters of about 3-4 hours.

Despite the possibility that impure plutonium may not be appreciably less risky to proliferation, the decontamination factor attribute, ‘DF’, will be scored from 0 to 1, based on the ratio of the power of the decontamination factor with 1 being no separation of plutonium (equation 16). Suppose the power of the decontamination factor is ‘p’, in 10^p .

$$DF_j = \frac{1}{p} \quad (14)$$

3.1.4.3 *Number of effluent streams*

The number of effluent streams is included due to the increased likelihood of diversion. Obviously, a nuclear fuel processing plant is expected to have stringent accountability standards, but more streams increase the chance of error. Additional streams might also imply additional labor, which could increase the chance of diversion by a laborer. The attribute for number of effluent streams, ‘ES’, is determined by the ratio of the number of effluent streams in the technology to 1, which would imply the best possible number of streams for this measure (equation 17).

$$ES_j = \frac{1}{Streams_j} \quad (15)$$

3.1.5 Economics

The economics of reprocessing are usually argued in a fashion as reprocessing vs. open fuel cycle [16]. Comparative economics between several reprocessing methods is not generally employed, nor is there sense in such a comparison without significant scaling experiments. The economics of a facility will depend mostly upon: Size of the buildings, shielding requirements, nature of combined processes or facilities, location, and the processes involved. However, when a reprocessing facility is constructed in the United States, the process is likely to be unproven on a commercial scale. Lack of experience also leads to lack of confidence in costs. Regardless of the method employed, the legal framework developed by the Nuclear Waste Policy Act establishes federal government ownership of spent fuel [83]. Additionally, the case has been made that the ‘cost’ of reprocessing is also deeply a function of ownership, and financing of such a facility could make use of historical government behavior of ‘sunk’ debt or could rely on the innovation of the private sector [100]. For these reasons, the reprocessing technologies will not be evaluated or compared based on economics.

3.1.6 Stage of Development

In the previous method, stage of development was scored from the conceptual to commercial scale, and this score became part of the overall score, or ‘performance factor’ [96]. Due to the sensitivity of technology to learning, including such a factor in the comparison of technologies appears to be contrary to the goal of determining the best applicable technology. Technologies such as reprocessing are assumed to operate under dynamic increasing returns. The literature on ‘learning’ provides insights into this issue.

“Technical change in general can be ascribed to experience, that it is the very activity of production which gives rise to problems for which favorable responses are selected over time” [4]. Essentially, if a technology is being used, it is evolving through trial and error. In addition, this learning often leads to an effect where “early use of one technology can create a snowballing effect by which that technology quickly becomes preferred to others and comes to dominate the market” [25]. This early advantage is not guaranteed to last, but the correct ‘best’ technology is described as the one “which, if it were to be the surviving one,

would maximize net benefits from the technology choice process” [25]. For these reasons, the stage of development will not be considered in this decision analysis; however, decision makers are likely to reinsert this attribute, at least implicitly, if selection is required under pressure to begin operations in the near term.

3.2 Overall Scoring

The idea of an overall score is necessary to allow for one evaluated reprocessing method to be selected as the ‘best’ choice based on the attributes described above. The overall score, or utility, ‘U’, of a technology is additive as shown in equation 4 of the Background. For each technology, ‘j’, there will be a utility, ‘U’, which is composed of the summation of the product of the weights, ω , and values, described above, for each attribute, ‘i’. The values then are the technical part of the utility and the weights provide for emotional, political, or social factors. Values are intended to be objective while weights are inherently subjective. In analysis, the weights, ω , for each attribute are initially assigned equivalence. In this base case, no preferences are analyzed. In the Discussion, different weighting options are described which show how preferences of the decision maker could influence the selected reprocessing technology. As previously mentioned in the Background, the summation of all ω must be one.

CHAPTER IV

REPROCESSING TECHNOLOGIES

Over the last 70 or so years, several methods of processing used nuclear fuel and other radioactive materials for separations have been proposed. For the purposes of this study, only those which are applicable to oxide fuels are discussed; the author does not wish to speculate on the possible future fuel types which could include: nitrides, carbides, or metals. Once a future fuel cycle has been determined, evaluation can be made for how to deal with the fuel; there remains sufficient spent oxide fuel (55,000 MTHM by the end of this year [14]) for a dedicated technology—especially considering that most studies on plant design have focused on capacities of less than 3,000 MTHM processed per year. Considering the 2,000 MTHM discharged nuclear fuel per year, the legacy and accumulating fuel would take more than 50 years to process, and the lifetime of a reprocessing facility may be less than 50 years.

The Nuclear Energy Agency's (NEA) working group on flow sheet studies is evaluating the following technologies as they apply to oxide fuels: PUREX (current), PUREX with additional separations, PUREX with different solvents, UREX+2, PYROX, Dimitrovgrad Dry Process, fluoride volatility, and the electrometallurgical process by the Central Research Institute of Electric Power Industry (CRIEPI) [78].

The presented analysis considers ten technologies, shown in table 1, which align well with those under study by the NEA. The technologies chosen had enough published works to be able to describe the process and apply the methodology; additionally, these technologies are all currently employed or under consideration by one or many nations. There are other technologies which may become developed to the point where they could also be included in such an analysis at a later date, such as the molten carbonate eutectic proposed by Griffiths [40]. The UREX+1, UREX+2, and FLUOREX processes are not separate technologies like the others; rather, they are collections of technologies which are considered as one

process. Any separations technology could be followed by additional extraction processes such as those already included as a part of these. For a brief but clear description of some additional extraction processes, which include TRUEX, DIAMEX [98], and TALSPEAK, among others, see the Nuclear Energy Agency report on Actinide Separation Chemistry [75]. The process flow sheets shown here are for clarification of major steps in the technologies as described in the literature; not all of these steps follow the linear progression implied by such a simplified drawing.

4.1 Process similarities

Since the scope of this study is only current oxide fuels and legacy fuels from light water reactors, the make up of the fuel fed into each technology is assumed to be the same. This simplification allows for the expectation that the fission product gases released during cladding removal and dissolution will be the same for all technologies. Also, each technology can utilize the same range of cladding removal methods and storage options. Regardless of the technology employed for separation, there are a couple of attributes which will not be affected: Cladding removal, Off-gas treatment, and Storage. These attributes will not be included in the analysis because engineering advances can effect them without regard for the technology used in the separations.

4.2 Aqueous Processes

The solvent extraction methods currently considered are all of the liquid-liquid type: PUREX, COEX, Supercritical CO₂, UREX, UREX+1 and UREX+2. These technologies are all based on the principles of hydrometallurgy, and are adaptations of the PUREX process to some extent.

4.2.1 PUREX

Plutonium URanium EXtraction, or PUREX, was first developed by Knolls Atomic Power Laboratory (KAPL) at Oak Ridge National Laboratory (ORNL) [31] as mentioned in the Background chapter. There are exhaustive explanations of the theory and history associated with solvent extraction by the PUREX process which need not be fully restated here; a

summary is included. The process is based around the selective affinity of tributyl phosphate (TBP) for uranium and plutonium. There are three basic process functions 1)Extracting, where uranium and plutonium are separated from fission products; 2)Partitioning, where plutonium is extracted from uranium; and 3)Stripping. The plutonium undergoes further decontamination, with an average decontamination factor greater than 10^6 [10, 18].

After the fuel is chopped and declad, fuel is dissolved in nitric acid to create a feed solution. This solution is then input to a solvent extraction process with TBP diluted in n-dodecane ($C_{12}H_{26}$). While n-dodecane and TBP are combustible, explosions are not likely unless ‘red oil’ is formed. Red oil is produced when TBP, dodecane, and nitric acid are combined at a temperature over 120 C; this combination can explosively decompose at temperatures greater than 130 C [92].

A flow sheet for the PUREX process, Figure 1, shows the eight major steps.

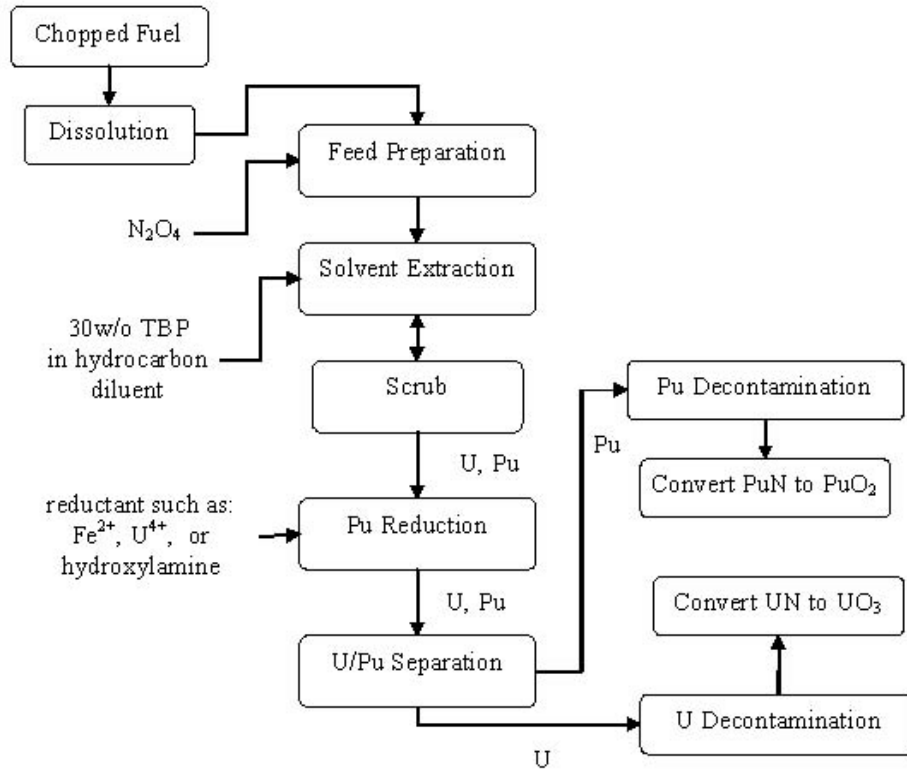


Figure 1: PUREX Flow Sheet, adapted from Benedict[10]

While PUREX is currently the most widely used technology with the greatest industrial history, there is considerable pressure to move away from this technology, and PUREX is not currently being considered under the Advanced Fuel Cycle Initiative [30]. The reasons are summarized by Abney (1997) [1],

The generation of copious quantities of high active fission product-bearing aqueous and non-aqueous wastes is one aspect of this process which has reduced its use at present. This process also requires large production sites and equipment for it to be performed on a production scale. The inherent ability of this process to generate highly purified plutonium and uranium fractions also contributes to nonproliferation issues not easily resolved in today's political climate.

While this statement appears to have several measures of preference regarding weights of the attributes, it is not a statement which will be used to summarily discard PUREX from this consideration. In fact, if the PUREX technology was to be ranked highest for a decision-makers utility function applied to the attributes included here, the actual utility would be much higher due to economic and learning issues which are not included in this analysis. In the Discussion, consideration will be made for factors leading to the selection of the PUREX process as a technology of choice.

4.2.2 COEX

COEX, or the COmbined EXtraction of plutonium and uranium, was proposed as a simplification of the PUREX technology which would be simpler and not separate plutonium by itself. The COEX process removes the most complicated steps in the PUREX process by not separating uranium and plutonium [111]. This process was developed on the premise that it didn't make sense to separate the uranium from the plutonium if they would be combined again when making mixed oxide fuel. The flow sheet proposed by Zabunoglu and Ozdemir(2005) was constructed with the idea that the process PUREX process is reduced to the co-decontamination followed by co-stripping, with 6 major steps, as shown in Figure 2.

The decontamination factor was shown to decrease as molarity of the scrub solution

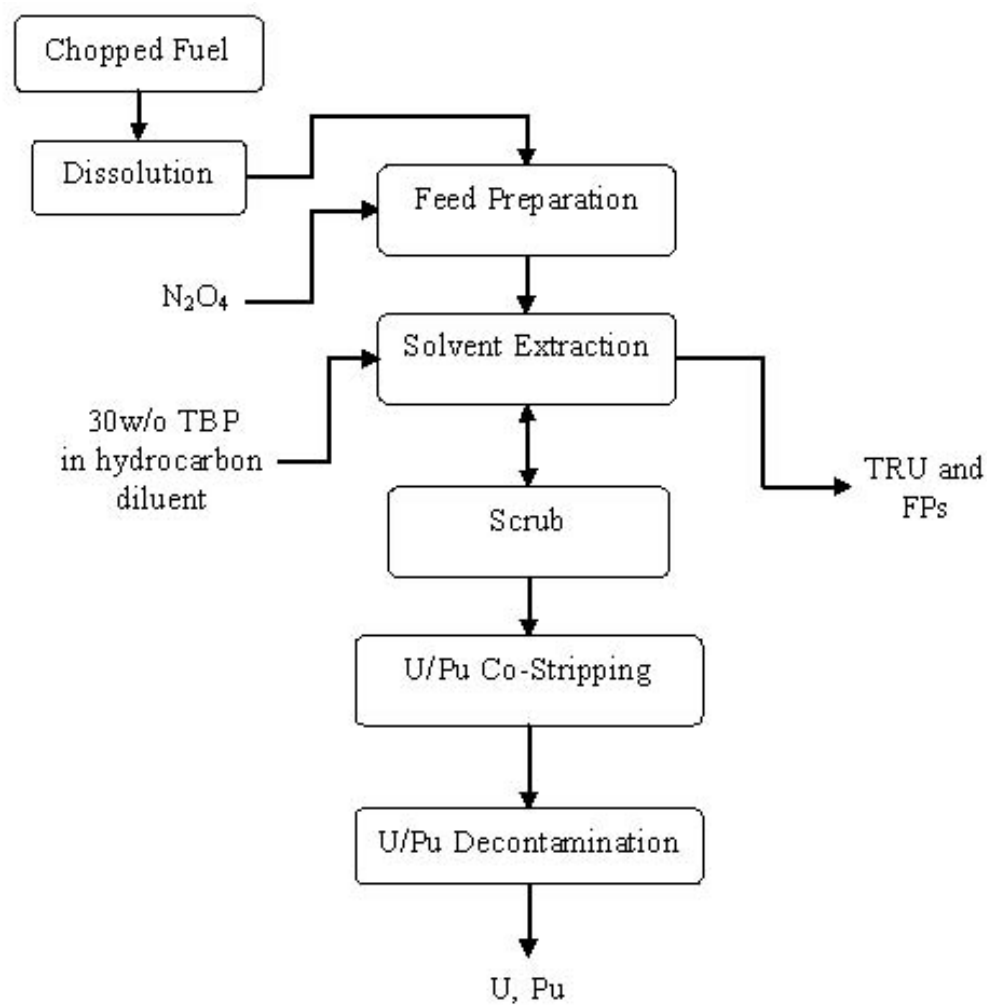


Figure 2: COEX Flow sheet [111]

increased, and the highest DF for rare earth fission products was on the order of 10^6 and for zirconium and ruthenium on the order of 10^3 . “The only product of purex co-processing is an aqueous solution of U+Pu, with a fissile content of approximately that of the SF solution (about 1.47 wt percent for SF from a typical LWR)” [111].

4.2.3 Supercritical CO_2

“Supercritical CO_2 can be used as a promising ecological alternative in liquid-liquid extraction systems” [95]. A supercritical fluid is defined by being above the critical temperature and pressure; for carbon dioxide, this is at 73.8 bar and 31.1 degrees C [33]. A solvent extraction method which employs Supercritical CO_2 with tributylphosphate has been shown to have similar characteristics to the PUREX technology it hopes to replace without the organic diluent. Experiments have shown that the Supercritical CO_2 extracts UO_2 as well as U_3O_8 [34]. The decontamination factor has been calculated on the order of 10^4 [107].

Experiments using this technology are ongoing, as this is the newest technology presented (and it is considered ‘greener’); here, the construction offered by Clifford (2001) is presented. $\text{UO}_2(\text{NO}_3)_2$ in 6M nitric acid is used to prepare U(VI). 0.01M U(VI) in 6M nitric acid is prepared in a liquid sample through which supercritical CO_2 under 200 bar (150,013 torr) flows [20]. Through this mixing, uranium is complexed and extracted to a chloroform solvent. The uranium in the solvent is then back extracted with $(\text{NH}_4)_2\text{SO}_4$. Neutron activation analysis in the Clifford (2001) study revealed that extraction efficiency decreases over time, but were comparable with those of traditional PUREX [20]. The process is composed of seven main steps as shown in Figure 3.

Experiments conducted by Japan Nuclear Cycle Development Institute showed that with the Joyo fuel, 16 to 40 grams at burn ups of 32000MWd/MTHM and 64,000MWd/MTHM could be extracted in 100 to 200 mL of a solution of TBP and 4.8mol/L nitric acid in less than 300 minutes [59]. They are calling this process, super-DIREX, for DIREct EXtraction due to the combination of dissolution and extraction steps. This particular form of the supercritical CO_2 technology does not have enough information to be considered separately.

This process has been modeled to be similarly effective under different complexes of

TBP and nitric acid; the presented data covered ratios of 0.7 and 1.8 TBP/nitric acid [35]. Researchers are still determining the underlying mechanisms for the observed behaviors of TBP in water and nitric acid solutions [95]. The process promises to decrease time, complexity, and liquid wastes over the current extraction technologies [59]

4.2.4 UREX

The URanium EXtraction, UREX, technology is defined by its “ability to separate out pure streams of uranium and technetium and nothing else” [15]. These separations are at efficiencies of ≥ 99.9 percent for uranium and >95 percent for technetium.

UREX basically uses the same process as PUREX, nitric acid solution with TBP and dodecane solvent, with the addition of acetohydroxamic acid (AHA) as a complexant in the scrub stage. AHA prevents the extraction of plutonium and neptunium and decomposes gaseously during evaporation (so addition of AHA does not contribute additional solid waste)[53].

The simplified process removes two steps from the PUREX process. The flow sheet developed by Rudisell (2003) for the case of application to the Dresden reactor fuel for experiment has been modified for the general case. The UREX process is composed of six steps as shown in Figure 4. Extraction and scrub stages are kept below 25 C to maintain Tc extraction [91]. While this requires addition of chill water, 25 C is 298.15 K which is very close to the temperatures used in off-gas treatment systems.

4.2.5 UREX+1

UREX+1 is one of a series of modifications to the UREX process. The difference between UREX and UREX+1 lies in the additional separation of a neptunium and plutonium stream in UREX+1. This Np/Pu stream could be used to create mixed oxide (MOX) fuel which can be burned in light water reactors. UREX+1 has been demonstrated on a laboratory scale in 2003 [90]. The UREX+1 process consists of eleven steps as shown in Figure 5.

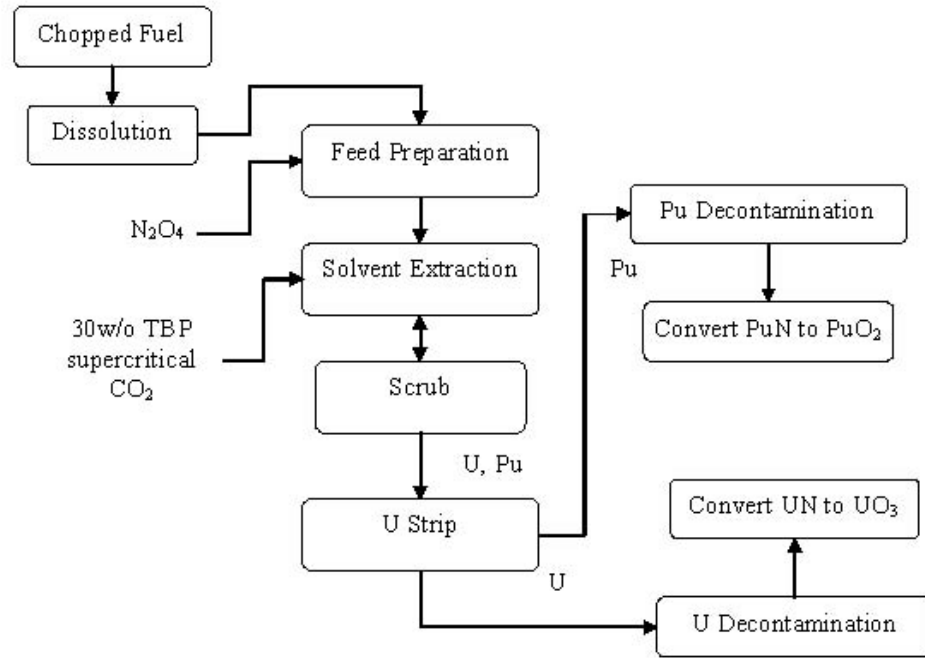


Figure 3: Supercritical CO₂ Flow Sheet, adapted from Clifford and Koyama [20, 59]

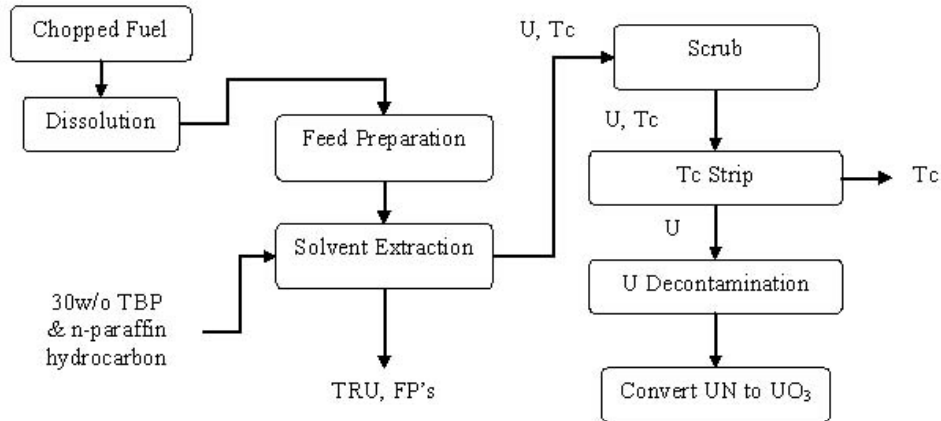


Figure 4: UREX Flow Sheet [91]

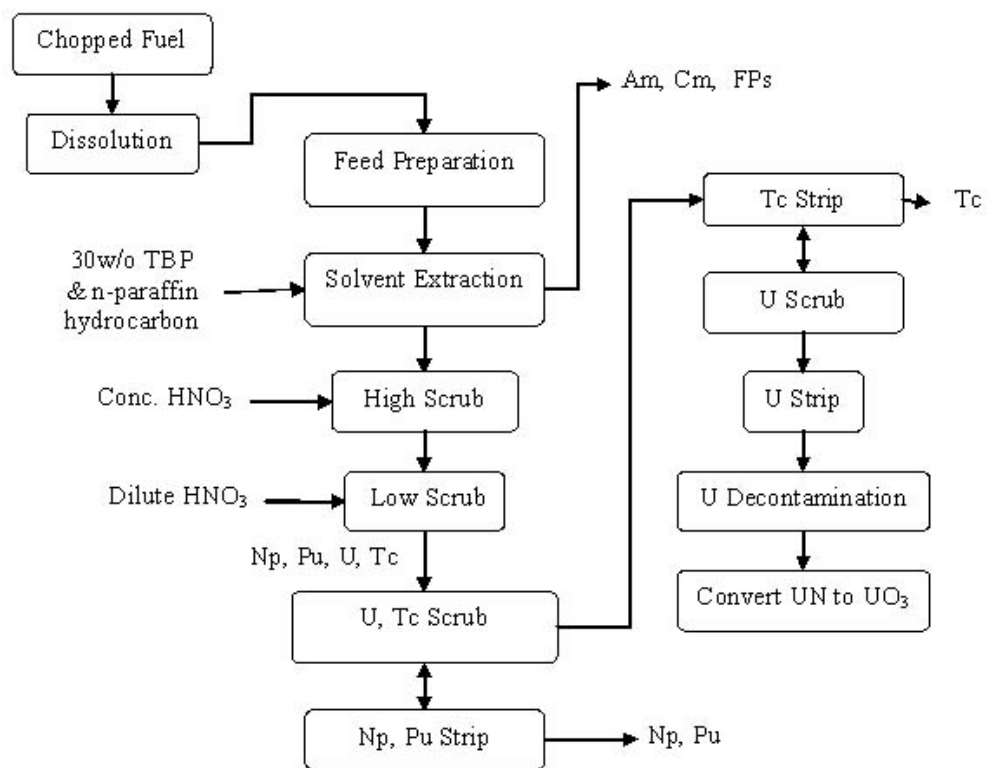


Figure 5: UREX+1 Flow Sheet [21]

4.2.6 UREX+2

UREX+2 is another in a series of modifications to the UREX process. UREX+2 uses ion exchange instead of stripping with concentrated nitric acid to separate technetium from uranium. Additionally, UREX+2 utilizes CCD-PEG and TALSPEAK processes. CCD-PEG is named for the extraction chemicals of chlorinated cobalt dicarbollide (CCD) and polyethylene glycol (PEG) which selectively extract cesium and strontium, respectively[67]. Separation of cesium and strontium from the fission product waste stream can reduce the short term heat load of the repository[30]. TALSPEAK is an acronym for ‘trivalent actinide lanthanide separations by phosphorus-reagent extraction from aqueous complexes’; this process was developed in the 1960’s and separates trivalent actinides from lanthanides and other fission products. The reported demonstration of the UREX+2 technology have not included experimental results on this portion of the proposed process [86]. The UREX+2 collection of technologies consists of fourteen steps as shown in Figure 6.

The UREX+1 and the UREX+2 processes are not the only adaptations that have been put forth by the Department of Energy. There is a suite of modified UREX processes, as shown in Table 4; however, the other processes have not been reported in the literature with enough information to add them to this comparison. In the future, such processes (combinations of processes) could be added to the decision by determining values for their attributes.

Table 4: Suite of UREX+ Processes [21, 22]

Process	Product Outputs						
Name	1	2	3	4	5	6	7
UREX	U	Tc	TRU+FPs				
UREX+1	U	Tc	Pu+Np	Am+Cm+FP			
UREX+1a	U	Tc	Cs/Sr	TRU	FP		
UREX+2	U	Tc	Cs/Sr	Pu+Np	Am+Cm+Ln	FP	
UREX+3	U	Tc	Cs/Sr	Pu+Np	Am+Cm	FP	
UREX+4	U	Tc	Cs/Sr	Pu+Np	Am	Cm	FP

4.3 Non-aqueous Processes

The non-aqueous processes include fluoride volatility, FLUOREX and pyroprocessing. The fluoride volatility process is classified as a physical separation; whereas, the pyroprocesses are classified as chemical extractions. The pyroprocessing technologies included in this comparison employ a molten salt electrorefining process as their main characteristic.

4.3.1 Fluoride volatility

Fluoride volatility takes advantage of the volatility of hexafluorides of uranium, neptunium, and plutonium in order to separate them from other materials as well as each other. After a rocky experimental start at Brookhaven National Laboratory due to unstable reactions, the process was reconfigured under the titles nitrofluor and aquafluor by future experiments [10]. Fluoride volatility has been shown to be most effective when used on fuels that contain little plutonium or neptunium because the fluorides PuF_6 and NpF_6 are less stable than UF_6 [75] (see Table 5).

Table 5: Sublimation points for AnF_6 [75]	
Compound	Sublimation/Boiling point (K)
UF_6	329.69
NpF_6	328.33
PuF_6	335.31

The decontamination factor for fluoride volatility is shown to be on the order of 10^5 with some fission product fluorides (of Nb, Tc, Mo, and Ru) which volatilize with the uranium fluoride [69]. A process flow sheet, Figure 7, shows the four steps included in this process.

A ‘hybrid recycle system’ has been proposed in Japan by Hitachi, Inc, where they suggest a slightly different approach. The fluoride volatility process is ‘improved’ to extract PuF_6 as well as UF_6 and remove the purification of these fluorides [50]. Previous studies as mentioned above suggest this may not be an improvement due to the variation in volatility of U and Pu.

4.3.2 FLUOREX

The FLUOREX technology comprises a collection of aqueous and non-aqueous processes which includes: AIROX process for the decladding, fluoride volatility, and PUREX (shown in Figure 8). AIROX is a process by which holes are drilled in the fuel cladding and then the fuel is subjected to cycling oxidation and reduction cycles; expansion of the U_3O_8 , which is formed by oxidizing UO_2 , causes the fuel to break apart [97]. The fluoride volatility process follows to remove the bulk of the uranium in a hexafluoride; this is followed by a partial PUREX process. The advantage of this technology is that the PUREX process steps can be conducted at approximately one-tenth of the scale that would be necessary if the uranium had not already been removed by fluoride volatility. This is the technology being pursued in Japan by Hitachi [36, 58].

4.3.3 Pyroprocessing

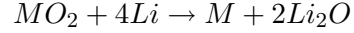
The five main steps for pyroprocessing, Figure 9, are the same for both presented technologies. Their variations will be discussed below. The general description of the process draws heavily on Ackerman, Laidler, McPheeters, and Stevens [2, 63, 71, 102].

Research in pyroprocessing has focused on the partitioning of metal fuels, such as would be found in a fast reactor. The oxide fuel of a light water reactor does not behave in the same way as metal fuel when immersed in the metal salt eutectic. As such, these pyroprocesses will not work directly on oxide fuel; however, methods to overcome this problem are already under demonstration and are not expected to be a limiting factor on implementing one of these technologies for oxide fuels. So, the first step following chopping of the fuel is oxide-reduction to metal or a conversion to chloride which can be accomplished in a number of ways, as shown below, and the method does not need to be specified so long that it accomplishes the reduction.

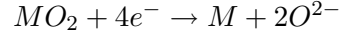
Examples of oxide reduction and conversion to chloride is shown by Sakamura (2004) [93]. For ‘M’ is U or Pu.

Reduction to Metal

Lithium Reduction



Electrochemical Reduction

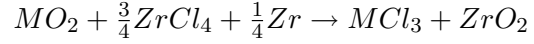


Conversion to Chloride

Chlorination using Cl_2 and carbon



Chlorination using $ZrCl_4$



Following reduction or chlorination, the fuel is placed in a basket in an electrorefiner (ER) which is a crucible containing the molten eutectic (chloride salts), a molten cadmium pool, and a solid cathode, and a liquid cathode; uranium is selectively electroplated onto the solid cathode [71] (see Figure 10). A smaller molten cadmium pool adds a second cathode which recovers remaining uranium, TRU, and a fraction of rare-earth fission products remaining. Most of the fission products collect in the chloride salt; the gaseous fission products: H_2 , Kr, and Xe are released and treated in off-gas; and the cladding hulls, Zr, and noble fission products accumulate in the cadmium pool or the basket. Where they go depends on their Gibbs Free Energy (see Table 6), $-\Delta G$, The eutectic and cadmium pool can be recycled to remove the accumulating contaminants [48].

Table 6: Gibbs Free Energies, at 500 degrees C [63, 60]

Stable Chlorides will remain in salt	Electrotransport	Unstable Chlorides will remain in metal
BaCl 87.2	CmCl3 64.0	CdCl2 32.2
CsCl 87.8	PuCl3 62.4	FeCl2 29.2
RbCl 87.0	AmCl3 62.1	NbCl5 26.7
KCl 86.7	NpCl3 58.1	MoCl4 16.8
SrCl2 84.7	UCl3 55.2	TcCl4 11.0
LiCl 82.5	ZrCl2 46.6	RhCl3 10.0
NaCl 81.2	PdCl2 9.0	
CaCl2 80.7	RuCl4 6.0	
LaCl3 70.2		
PrCl3 69.0		
CeCl3 68.6		
NdCl3 67.9		
YCl3 65.1		

There are still questions regarding ways in which to improve separations as research

shows incomplete separation of actinides and lanthanides from these processes [64]. However, follow on treatments which are capable of An/Ln separation can be applied and are not discussed here.

4.3.3.1 *LiCl-KCl*

A brief history of the development of this processes was presented by Benedict (2002) [11], and it will be summarized here. In the 1980's, Argonne National Laboratory (ANL) began development of an electrometallurgical process for the treatment of spent fuel from Experimental Breeder Reactor-II (EBR-II) in the early 1980's. The technology was demonstrated between 1996 and 1999 with final evaluation by the National Academy of Science in 2000. In September of 2000, the technology was implemented for treatment of EBR-II fuel, following completed Environmental Impact Statement and completed Spent Fuel Treatment Implementation Plan. The process for separations in the LiCl-KCl process is the same as that shown for the general pyroprocess with LiCl-KCl being the eutectic.

There are still fundamental questions relating to the behavior of materials in the eutectic. Kuznetsov discusses the inconsistencies in previous experiments which used oxide materials within the set-up. He goes on to determine that the Distribution Coefficients are dynamic with temperature and should be examined in more depth [61].

4.3.3.2 *NaCl-KCl*

While the United States researchers have focused primarily on the lithium chloride eutectic, there has been significant progress in eastern Europe on a sodium chloride eutectic. The bulk of this research is in conjunction with a specific technology for vibration packaged fuel which will not be discussed. This process is very similar to the lithium chloride method with a NaCl-KCl eutectic [75]. The differences for the sodium chloride process is that it is expected to operate at a higher temperature, 1000 K, compared to 833 K max for the LiCl process.

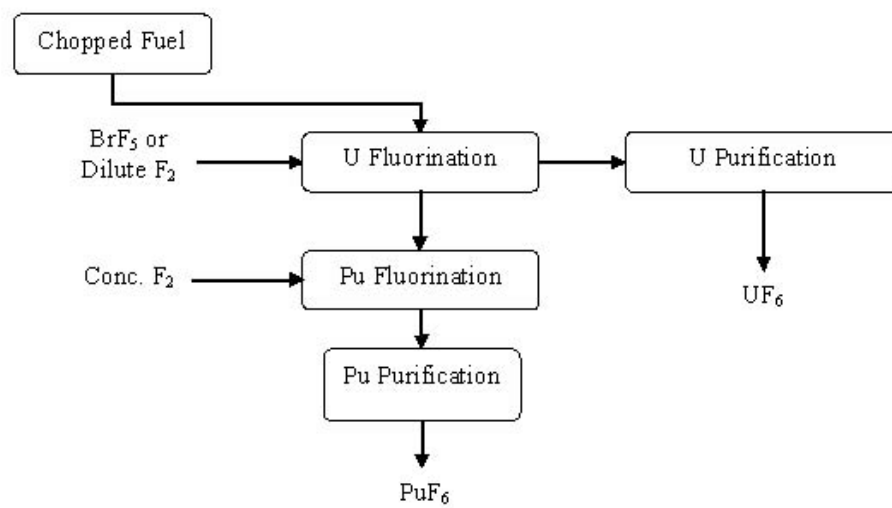


Figure 7: Fluoride volatility flow sheet [50]

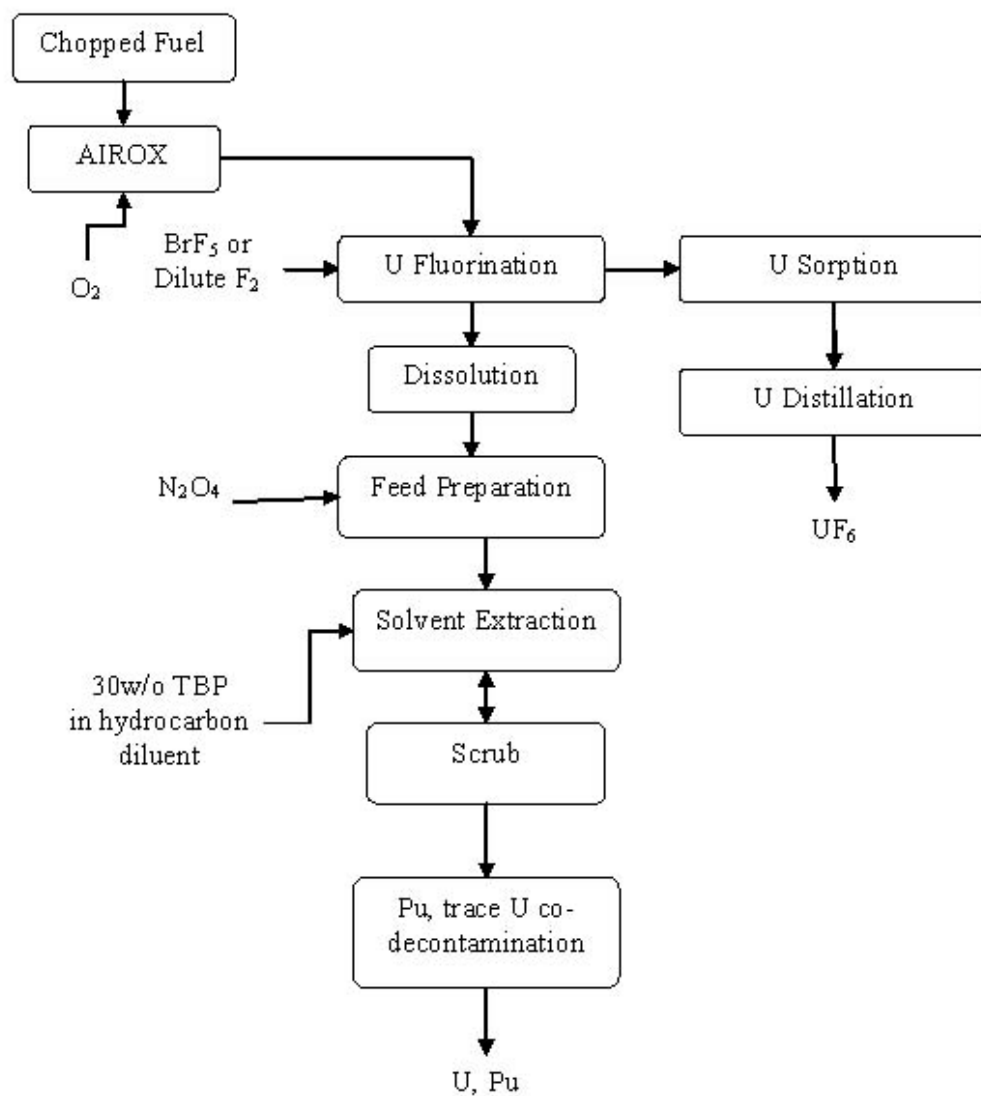


Figure 8: FLUOREX Process Flow Sheet [10, 50, 36]

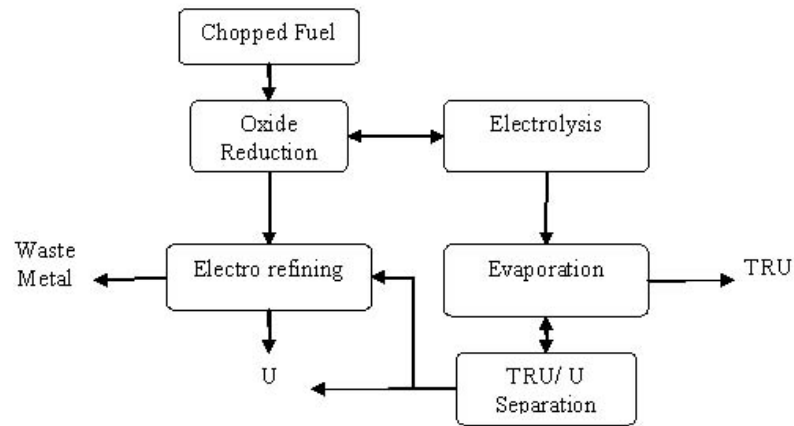


Figure 9: Pyroprocess Flow Sheet [19, 71]

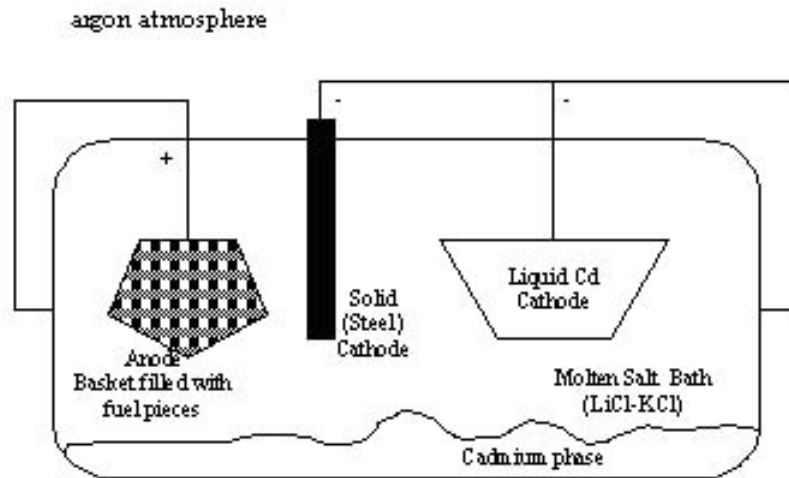


Figure 10: Representation of Electrorefiner [64, 19]

CHAPTER V

APPLICATION OF METHOD AND RESULTS

The application of the multiattribute decision analysis method is presented through each attribute. Because the details of the technologies and the decision analysis methodology have already been discussed in detail in previous chapters, tables are provided for ease of comparison.

5.1 *Attribute Scores*

This section is set up as a series of tables which present the data and scores for each technology on each attribute. The attribute data are taken from the same sources listed in the Technology description chapter. Attribute scores are rounded to five decimal places.

5.1.1 Number of Steps

The number of steps, shown in Table 7, are determined through a count of steps as described in the technologies chapter. The numbers in the table correspond with the steps in the related flow sheet.

Table 7: Number of steps, by technology		
Technology	Number of Steps	NS
PUREX	8	0.12500
COEX	6	0.16667
Supercritical CO ₂	7	0.14286
UREX	6	0.16667
UREX+1	11	0.09091
UREX+2	14	0.07143
Fluoride Volatility	4	0.25000
FLUOREX	9	0.11111
LiCl-KCl	5	0.20000
NaCl-KCl	5	0.20000

5.1.2 Operating Temperature

The operating temperature, shown in Table 8, is a minimum or maximum, as described in the technologies chapter.

Table 8: Operating temperature, by technology

Technology	Max/Min Operating Temperature (K)	OT
PUREX	ambient	1.00000
COEX	ambient	1.00000
Supercritical CO ₂	ambient	1.00000
UREX	298.15	0.79901
UREX+1	298.15	0.79901
UREX+2	298.15	0.79901
Fluoride Volatility	775	0.48148
FLUOREX	775	0.48148
LiCl-KCl	833	0.44796
NaCl-KCl	1000	0.37315

5.1.3 Operating Pressure

The operating pressure, shown in Table 9, is a minimum or maximum as described in the technologies chapter.

Table 9: Operating Pressure, by technology

Technology	Max/Min Operating Pressure (torr)	OT
PUREX	ambient	1.00000
COEX	ambient	1.00000
Supercritical CO ₂	105,103	0.00714
UREX	ambient	1.00000
UREX+1	ambient	1.00000
UREX+2	ambient	1.00000
Fluoride Volatility	ambient	1.00000
FLUOREX	ambient	1.00000
LiCl-KCl	0.1	0.00013
NaCl-KCl	0.1	0.00013

5.1.4 Use of Corrosive Materials

Scoring for the use of corrosive materials is shown in Table 10.

Table 10: Use of corrosive materials, by technology

Technology	Corrosive?	CM
PUREX	Nitric Acid	0.50000
COEX	Nitric Acid	0.50000
Supercritical CO ₂	Nitric Acid	0.50000
UREX	Nitric Acid	0.50000
UREX+1	Yes	0.10000
UREX+2	Yes	0.10000
Fluoride Volatility	Yes	0.10000
FLUOREX	Yes	0.10000
LiCl-KCl	Yes	0.10000
NaCl-KCl	Yes	0.10000

5.1.5 Maximum Credible Accident

The total toxicities for the maximum credible accident (Table 11) come from the analysis by Selvaduray (1978) [96], as stated in the methodology. For those technologies that were not described in the previous work, the value from the technology which is most similar is applied; this holds because the release is primarily a function of the materials used (i.e. liquid solvent, chloride eutectic). Also, I have assumed that FLUOREX has toxicity as the summation of that from Fluoride volatility and a tenth of that from the other aqueous processes due to lower volume in the aqueous process.

Table 11: Maximum Credible Accident Toxicity, by technology

Technology	Toxicity	MCA
PUREX	7.1×10^9	0.14085
COEX	7.1×10^9	0.14085
Supercritical CO ₂	7.1×10^9	0.14085
UREX	7.1×10^9	0.14085
UREX+1	7.1×10^9	0.14085
UREX+2	7.1×10^9	0.14085
Fluoride Volatility	3.45×10^9	0.28986
FLUOREX	4.16×10^9	0.24038
LiCl-KCl	4.8×10^9	0.20833
NaCl-KCl	4.8×10^9	0.20833

5.1.6 Explosiveness

True or false scoring for explosiveness is given in Table 12. The explosiveness of a process can be argued. For example, lithium is quite reactive with water and oxygen, but it does

not exist in its pure state due to this behavior. Sodium is even more reactive. However, the chloride form for both appears stable under given conditions.

Table 12: Explosiveness, by technology

Technology	Explosive?	EX
PUREX	Yes	0
COEX	Yes	0
Supercritical CO ₂	No	1
UREX	Yes	0
UREX+1	Yes	0
UREX+2	Yes	0
Fluoride volatility	No	1
FLUOREX	Yes	0
LiCl-KCl	No	1
NaCl-KCl	No	1

5.1.7 Secondary Waste

Scoring for secondary waste generation is shown in Table 13. In determining secondary waste volume, some technologies did not have data which were congruent enough to determine the volumes. For those technologies, COEX, UREX, UREX +1 and UREX+2, the assumption has been made that they will not produce more waste than the technology that they are supposed to be improving upon. The UREX+ processes are designed to not produce high level liquid wastes [22]. Supercritical CO₂ values were determined as a ratio from PUREX values based on estimates in the literature.

Table 13: Secondary Waste Generated, by technology

Technology	Secondary Waste Volume m ³ per kgHM	Liquid/Total	SW
PUREX	0.38498	1	0.01323
COEX	Not Stated	1	0.01323
Supercritical CO ₂	0.32082	1	0.09441
UREX	Not Stated	1	0.01323
UREX+1	Not Stated	1	0.01323
UREX+2	Not Stated	1	0.01323
Fluoride volatility	0.00105	0.52381	0.73677
FLUOREX	0.39550	0.98736	0.00632
LiCl-KCl	0.45000	0	0.99997
NaCl-KCl	0.45000	0	0.99997

5.1.8 Separate plutonium stream

Table 14 shows the scoring for the existence, or lack of, a separate plutonium stream.

Table 14: Separated plutonium stream, by technology

Technology	Separate plutonium?	SPu
PUREX	Yes	0
COEX	No	1
Supercritical CO ₂	Yes	0
UREX	No	1
UREX+1	No	1
UREX+2	No	1
Fluoride volatility	Yes	0
FLUOREX	Yes	0
LiCl-KCl	No	1
NaCl-KCl	No	1

5.1.9 Decontamination Factor

The decontamination factors were described in the technologies chapter; table 15 shows the decontamination factors and related scores.

Table 15: Decontamination Factor, by technology

Technology	Decontamination Factor	DF
PUREX	$10^6 - 10^8$	0.12500 - 0.16667
COEX	$10^3 - 10^6$	0.16667 - 0.33333
Supercritical CO ₂	10^4	0.25000
UREX	10^6	0.16667
UREX+1	$10^4 - 10^7$	0.14286 - 0.16667
UREX+2	$10^4 - 10^7$	0.14286 - 0.16667
Fluoride volatility	10^5	0.20000
FLUOREX	10^7	0.14286
LiCl-KCl	10^5	0.20000
NaCl-KCl	10^5	0.20000

5.1.10 Number of Effluent Streams

The number of effluent streams is simply a count of the different outputs from the process as can be seen by the process flowcharts. Table 16 shows the number of effluent streams.

Table 16: Number of effluent streams, by technology

Technology	Number of effluent streams	ES
PUREX	3	0.33333
COEX	2	0.50000
Supercritical CO-2	3	0.33333
UREX	3	0.33333
UREX+1	5	0.20000
UREX+2	6	0.16667
Fluoride Volatility	3	0.33333
FLUOREX	3	0.33333
LiCl-KCl	3	0.33333
NaCl-KCl	3	0.33333

5.2 Overall Scores

Recall, the overall score is calculated from the attribute scores and the weights as given in Equation 4 (Background). Each technology will have its own overall score, U . When assuming equal weight, the ω_i , for each i is set equal to 0.1 (there are ten attributes); overall scores are given in Table 17.

This equal weighting calculation shows that the COEX process with a $U=0.45464$ would be selected followed by fluoride volatility process at $U=0.43916$.

Table 17: Overall Scores with equal weight, by technology

Technology	NS	OT	OP	CM	MCA	EX	SW
PUREX	0.12500	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
COEX	0.16667	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
Supercritical CO ₂	0.14286	1.00000	0.00714	0.50000	0.14085	1.00000	0.14353
UREX	0.16667	0.79901	1.00000	0.50000	0.14085	0.00000	0.07224
UREX+1	0.09091	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
UREX+2	0.07143	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
Fluoride Volatility	0.25000	0.48148	1.00000	0.10000	0.28986	1.00000	0.73693
FLUOREX	0.11111	0.48148	1.00000	0.10000	0.24038	0.00000	0.06688
LiCl-KCl	0.20000	0.44796	0.00013	0.10000	0.20833	1.00000	0.50000
NaCl-KCl	0.20000	0.37315	0.00013	0.10000	0.20833	1.00000	0.50000
Technology	SPu	DF	ES	U			
PUREX	0.00000	0.12500	0.33333	0.32964			
COEX	1.00000	0.16667	0.50000	0.45464			
Supercritical CO ₂	0.00000	0.25000	0.33333	0.35177			
UREX	1.00000	0.16667	0.33333	0.41788			
UREX+1	1.00000	0.14286	0.20000	0.35459			
UREX+2	1.00000	0.14286	0.16667	0.34930			
Fluoride Volatility	0.00000	0.20000	0.33333	0.43916			
FLUOREX	0.00000	0.14286	0.33333	0.24761			
LiCl-KCl	1.00000	0.20000	0.33333	0.39898			
NaCl-KCl	1.00000	0.20000	0.33333	0.39150			

CHAPTER VI

DISCUSSION

From the results, in the absence of preferences, or in the face of equal preferences, the COEX technology would be chosen, assuming that the factors included as attributes in this analysis are considered comprehensive by the decision maker.

6.1 Additional Weighting Analysis

We could instead assume that a decision maker has a preference for low operating temperatures, but is not concerned at all with number of steps involved. For this analysis, the weights will be set such that $\omega_{OT} = 0.5$, $\omega_{NS} = 0.0$, and the other eight attributes all have weights of 0.0625 (Table 18).

Table 18: Overall Scores with preference for ambient operating temperature, by technology

Technology	NS	OT	OP	CM	MCA	EX	SW
PUREX	0.12500	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
COEX	0.16667	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
Supercritical CO ₂	0.14286	1.00000	0.00714	0.50000	0.14085	1.00000	0.14353
UREX	0.16667	0.79901	1.00000	0.50000	0.14085	0.00000	0.07224
UREX+1	0.09091	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
UREX+2	0.07143	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
Fluoride Volatility	0.25000	0.48148	1.00000	0.10000	0.28986	1.00000	0.73693
FLUOREX	0.11111	0.48148	1.00000	0.10000	0.24038	0.00000	0.06688
LiCl-KCl	0.20000	0.44796	0.00013	0.10000	0.20833	1.00000	0.50000
NaCl-KCl	0.20000	0.37315	0.00013	0.10000	0.20833	1.00000	0.50000
Technology	SPu	DF	ES	U			
PUREX	0.00000	0.12500	0.33333	0.63571			
COEX	1.00000	0.16667	0.50000	0.71123			
Supercritical CO ₂	0.00000	0.25000	0.33333	0.64843			
UREX	1.00000	0.16667	0.33333	0.60032			
UREX+1	1.00000	0.14286	0.20000	0.56550			
UREX+2	1.00000	0.14286	0.16667	0.56342			
Fluoride Volatility	0.00000	0.20000	0.33333	0.46950			
FLUOREX	0.00000	0.14286	0.33333	0.35846			
LiCl-KCl	1.00000	0.20000	0.33333	0.43284			
NaCl-KCl	1.00000	0.20000	0.33333	0.39544			

With a decision maker who strongly desires ambient operating conditions without concern for number of steps, the chosen technology is COEX with a $U=0.71123$, followed by Supercritical CO_2 and PUREX.

Another example shows a decision maker with a strong preference for proliferation resistance who thinks that a low Decontamination Factor and few Effluent Streams would be best for minimizing risk, such that ω_{DF} and ω_{ES} both = 0.3 and all other weights are 0.04444 (Table 19).

Table 19: Overall Scores with preference for proliferation resistance, by technology

Technology	NS	OT	OP	CM	MCA	EX	SW
PUREX	0.12500	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
COEX	0.16667	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
Supercritical CO_2	0.14286	1.00000	0.00714	0.50000	0.14085	1.00000	0.14353
UREX	0.16667	0.79901	1.00000	0.50000	0.14085	0.00000	0.07224
UREX+1	0.09091	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
UREX+2	0.07143	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
Fluoride Volatility	0.25000	0.48148	1.00000	0.10000	0.28986	1.00000	0.73693
FLUOREX	0.11111	0.48148	1.00000	0.10000	0.24038	0.00000	0.06688
LiCl-KCl	0.20000	0.44796	0.00013	0.10000	0.20833	1.00000	0.50000
NaCl-KCl	0.20000	0.37315	0.00013	0.10000	0.20833	1.00000	0.50000
Technology	SPu	DF	ES	U			
PUREX	0.00000	0.12500	0.33333	0.26364			
COEX	1.00000	0.16667	0.50000	0.37243			
Supercritical CO_2	0.00000	0.25000	0.33333	0.30542			
UREX	1.00000	0.16667	0.33333	0.31350			
UREX+1	1.00000	0.14286	0.20000	0.24521			
UREX+2	1.00000	0.14286	0.16667	0.23435			
Fluoride Volatility	0.00000	0.20000	0.33333	0.33148			
FLUOREX	0.00000	0.14286	0.33333	0.23174			
LiCl-KCl	1.00000	0.20000	0.33333	0.31362			
NaCl-KCl	1.00000	0.20000	0.33333	0.31029			

This decision maker would choose choose COEX at $U=0.37243$ followed by fluoride volatility, as in the no weighting case; however, there is less difference between the overall scores in this case.

In contrast, there are advantages to separating more streams for different effects on waste management and possible reuse of fuel. Assume a decision maker has a strong preference for more effluent streams, such that $\omega_{ES} = 0.5$ and the other nine attributes have weights of 0.0555. Additionally, the attribute for effluent streams in this case is $ES' = 1 - ES$ to

reverse the scoring such that more streams result in a higher score (see Table 20). The result is selection of the UREX+2 process followed by UREX+1 and fluoride volatility.

Table 20: Overall Scores with preference for many effluent streams, by technology

Technology	NS	OT	OP	CM	MCA	EX	SW
PUREX	0.12500	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
COEX	0.16667	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
Supercritical CO ₂	0.14286	1.00000	0.00714	0.50000	0.14085	1.00000	0.14353
UREX	0.16667	0.79901	1.00000	0.50000	0.14085	0.00000	0.07224
UREX+1	0.09091	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
UREX+2	0.07143	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
Fluoride Volatility	0.25000	0.48148	1.00000	0.10000	0.28986	1.00000	0.73693
FLUOREX	0.11111	0.48148	1.00000	0.10000	0.24038	0.00000	0.06688
LiCl-KCl	0.20000	0.44796	0.00013	0.10000	0.20833	1.00000	0.50000
NaCl-KCl	0.20000	0.37315	0.00013	0.10000	0.20833	1.00000	0.50000
Technology	SPu	DF	ES	U			
PUREX	0.00000	0.12500	0.33333	0.49795			
COEX	1.00000	0.16667	0.50000	0.47480			
Supercritical CO ₂	0.00000	0.25000	0.33333	0.51024			
UREX	1.00000	0.16667	0.33333	0.54697			
UREX+1	1.00000	0.14286	0.20000	0.58588			
UREX+2	1.00000	0.14286	0.16667	0.60147			
Fluoride Volatility	0.00000	0.20000	0.33333	0.55879			
FLUOREX	0.00000	0.14286	0.33333	0.45237			
LiCl-KCl	1.00000	0.20000	0.33333	0.53647			
NaCl-KCl	1.00000	0.20000	0.33333	0.53231			

A final example will assume that a decision maker needs to separate plutonium with a high decontamination factor and is not concerned greatly with other attributes. To construct the output, $\omega_{SPu'}$ and $\omega_{DF'}$ will be set to 0.4 each while the other eight weights are 0.02222. Like in the previous case, $SPu' = 1 - SPu$ and $DF' = 1 - DF$ to reverse the scoring logic. The resulting choice is PUREX followed by fluoride volatility (Table 21). Note that this is not exactly the decision faced by those who went forward with the PUREX process because the competing technologies have changed.

While examining discrete possible decision makers provides for some insight, such examination becomes an exercise without application when actual decision maker preferences are unknown. To understand fully the spectrum of outcomes, analysis must be made over a range of possibilities.

Table 21: Overall Scores with preference for separating plutonium and high decontamination factors, by technology

Technology	NS	OT	OP	CM	MCA	EX	SW
PUREX	0.12500	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
COEX	0.16667	1.00000	1.00000	0.50000	0.14085	0.00000	0.07224
Supercritical CO ₂	0.14286	1.00000	0.00714	0.50000	0.14085	1.00000	0.14353
UREX	0.16667	0.79901	1.00000	0.50000	0.14085	0.00000	0.07224
UREX+1	0.09091	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
UREX+2	0.07143	0.79901	1.00000	0.10000	0.14085	0.00000	0.07224
Fluoride Volatility	0.25000	0.48148	1.00000	0.10000	0.28986	1.00000	0.73693
FLUOREX	0.11111	0.48148	1.00000	0.10000	0.24038	0.00000	0.06688
LiCl-KCl	0.20000	0.44796	0.00013	0.10000	0.20833	1.00000	0.50000
NaCl-KCl	0.20000	0.37315	0.00013	0.10000	0.20833	1.00000	0.50000
Technology	SPu	DF	ES	U			
PUREX	0.00000	0.12500	0.33333	0.82048			
COEX	1.00000	0.16667	0.50000	0.40844			
Supercritical CO ₂	0.00000	0.25000	0.33333	0.77262			
UREX	1.00000	0.16667	0.33333	0.40027			
UREX+1	1.00000	0.14286	0.20000	0.39626			
UREX+2	1.00000	0.14286	0.16667	0.39508			
Fluoride Volatility	0.00000	0.20000	0.33333	0.81315			
FLUOREX	0.00000	0.14286	0.33333	0.79470			
LiCl-KCl	1.00000	0.20000	0.33333	0.38199			
NaCl-KCl	1.00000	0.20000	0.33333	0.38033			

6.2 Monte Carlo Analysis

Monte Carlo analysis allows for the consideration of the range of possibilities in a case of uncertainty. For this analysis, the technological attributes are taken as a given and the utility for decision makers for each attribute, or the weights, is uncertain. In order to analyze the multitude of possibilities, a model of the decision methodology was built in Decision Programming Language (DPL 6.0), and then a Monte Carlo analysis was run.

6.2.1 Model in Decision Programming Language

The model (Figure 11) is built with technologies as the decision based on the attributes and weights which form the overall score. The decision shows up as a square, calculations and constants shows up as rounded rectangles, and continuous chances (Monte Carlo) are represented by ovals. The decontamination factor is continuous chance with a uniform distribution (flat line representing equal chance for any value) for those technologies which had a range of decontamination factors presented in the literature. Each weight is also continuous chance; weights are ranged from 0 to 1 as a triangular distribution with a max at 0.1 (no preference).

6.2.2 Results

When the Monte Carlo analysis is run, the uncertainty in the decontamination factor (ranges) and in the weights (distributions) is analyzed by rolling the dice for each of these nodes. For each technology, DPL runs through the decision tree (Figure 12); at each chance node, dice are rolled one million times before moving to the next node.

The result is the overlapping curves showing where the technologies are likely to score. The high degree of overlap shows that the results are similar enough that no one technology completely dominates or is dominated, see Figure 13.

6.3 Sensitivity Analysis

The model in the decision software, DPL, also allows for sensitivity analysis. Sensitivity analysis is very important when making decisions with uncertainty. If changing a value causes a different outcome, it is imperative that the true value, or best approximation,

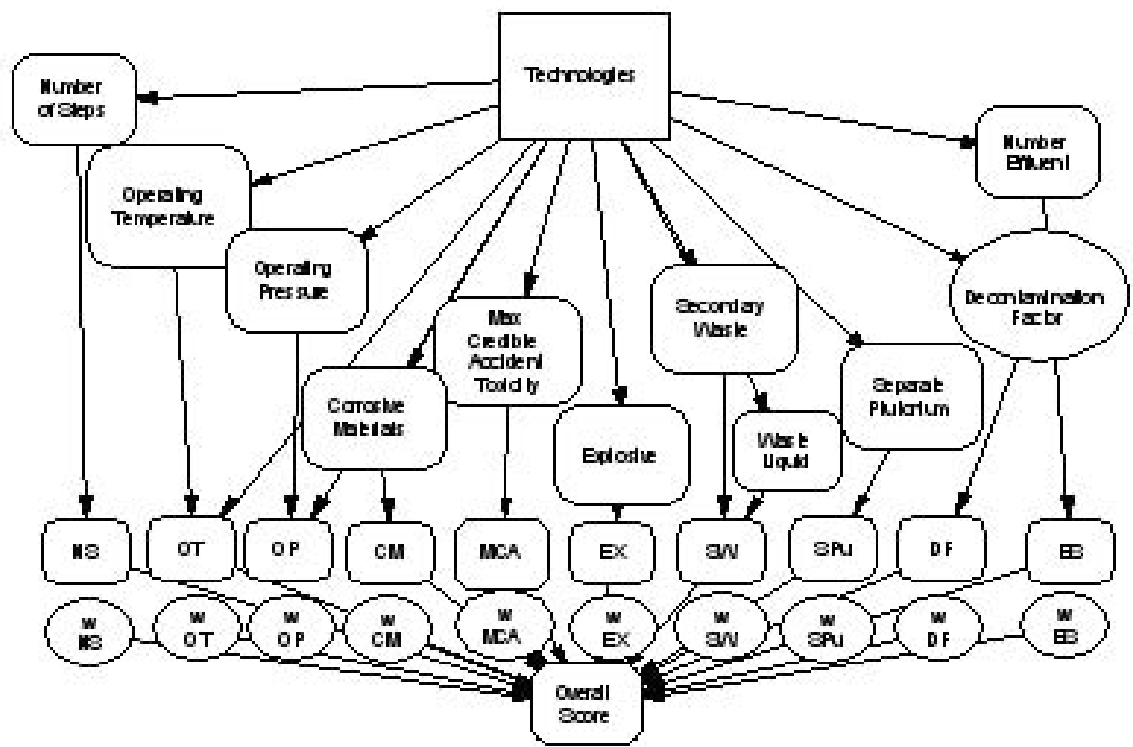


Figure 11: Model in DPL

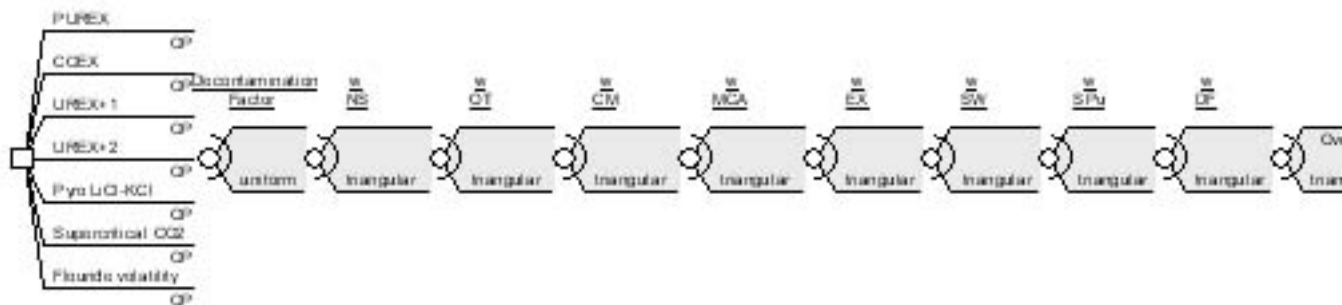


Figure 12: Decision Tree in DPL

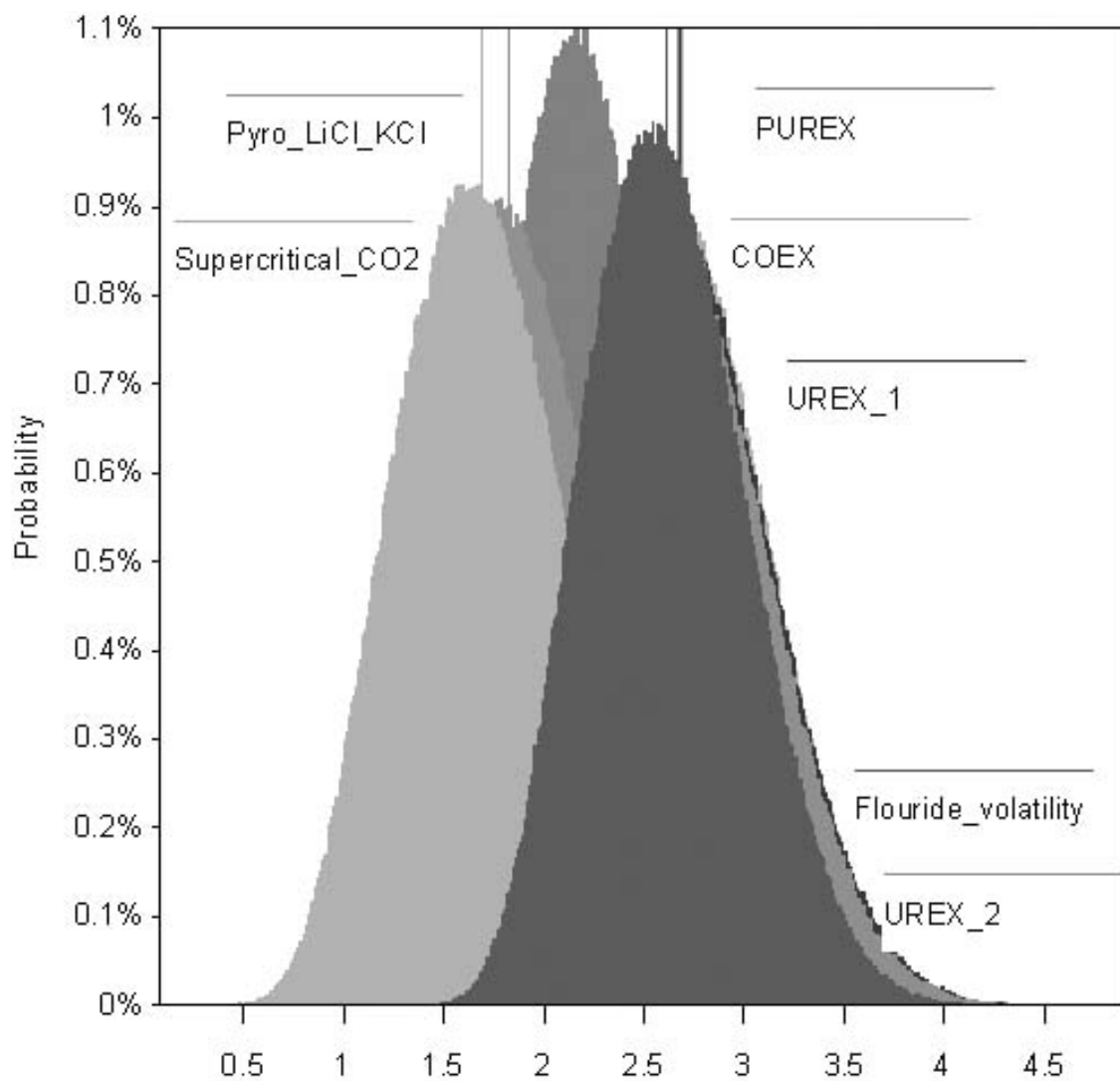


Figure 13: Monte Carlo Result

of that value be known. Sensitivity analysis can also inform the decision maker when a variable could be ignored. Variables which do not alter the outcome, when changed within reasonable limits, can be ignored to simplify the decision.

DPL displays sensitivity data in a tornado diagram (Figure 14). To generate this diagram, the values which are to be analyzed are selected at a 'base' value and allowed to vary between selected endpoints while holding all other variables at their 'base' value. The white area shows that there is no change from the 'best' choice from the base values when the weight in question is varied. A change in color shows that a different 'best' choice will exist once the weight in question reaches the point where the color change begins. Sensitivity analysis does not show what the new best choice will be; it only shows that varying weights can change the outcome, some more than others.

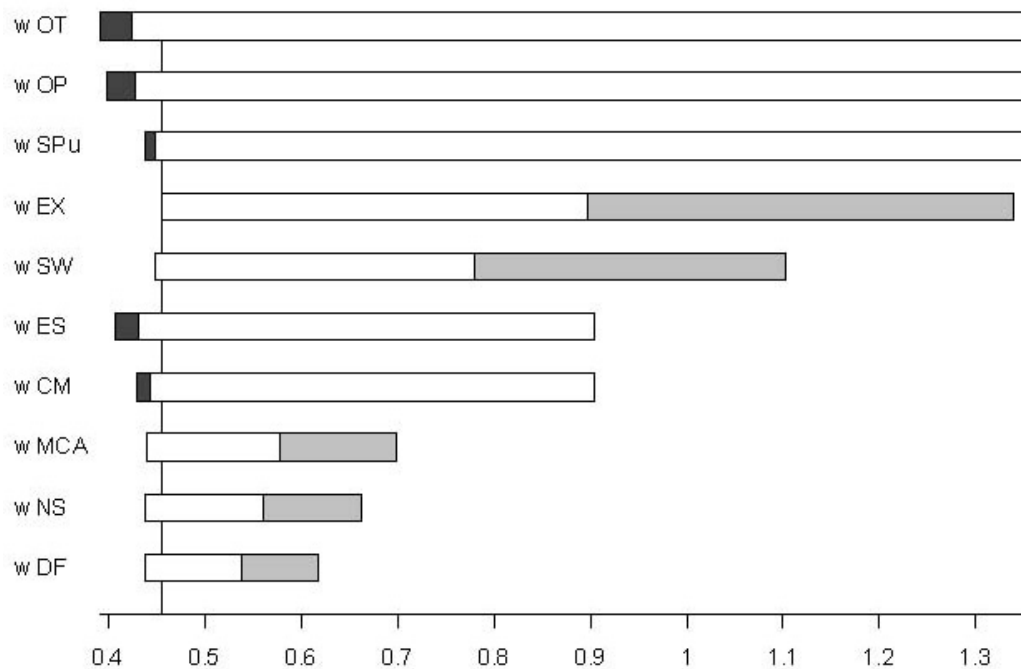


Figure 14: Sensitivity Analysis for Weights

6.4 Future Work

Future work should determine actual preferences. Determination of actual preferences of decision makers could prove difficult for an outside researcher; however surveys of decision

makers could be useful if a proxy for the attributes is used so that the decision maker would clearly understand the meaning of the attribute terminology. Additionally, historical analysis of preferences in time periods where decisions were previously made on a processing technology could prove useful. An example was shown above where the most important attribute is separation of plutonium; in this case, the presented methodology for ‘SPu’ and ‘DF’ is inverted. In such an analysis, PUREX scores better than other technologies, but these technologies are different from those which actually competed with PUREX at the time. This is a simplistic example of how historical analysis could be used to investigate the robustness of the methodology. Historical analysis could also offer improvements and additions to the methodology’s attributes.

CHAPTER VII

CONCLUSIONS

In this work, a decision methodology based on Multiattribute Utility Theory for application to a reprocessing technology choice was presented. The methodology examines ten attributes applicable to reprocessing; each attribute is scored, and each can be assigned a particular utility weight, ω . When no preferences are stated, the technology chosen is the COEX technology, followed by fluoride volatility. When preferences are introduced for operating temperature and proliferation the top choice remains COEX (with changes in the second ranking choice and differences in overall scores). When preferences are stated for more effluent streams, the choice becomes UREX+2. Additionally, in a case where it is desired to separate plutonium and have high decontamination factor, the PUREX technology is chosen. These are examples of how a decision maker might apply their utility function for reprocessing technology attributes to this methodology.

Due to the imposed circumstances put forth by the U.S. Congress [23], a decision must be made by the Department of Energy for a reprocessing path forward by next year. This is one attempt to organize the decision making process; improvements and additions can be made to this model. More work is necessary in this area to ensure that technological lock-in is not forced by undeveloped, possibly superior, technologies and uninformed decision makers [25].

REFERENCES

- [1] ABNEY, K. D., AVENS, L. R., ELLER, P. G., and ARTHUR, E. D., “Advanced nuclear fuel processing options final report,” tech. rep., Department of Energy, Los Alamos National Laboratory (LANL), 1997. LA-UR-98-2773.
- [2] ACKERMAN, J. P., “Chemical basis for pyrochemical reprocessing nuclear fuel,” *Industrial and Engineering Chemistry Research*, vol. 30, pp. 141–145, 1991.
- [3] ALFONSO, C. A. M. and CRESPO, J. G., eds., *Green Separation Processes: Fundamentals and Applications*. Weinheim, Germany: Wiley-VCH, 2005.
- [4] ARROW, K., “The economic implications of learning by doing,” *The Review of Economic Studies*, vol. 29, no. 3, pp. 155–173, 1962.
- [5] AVS, “Reference guide.” electronic glossary, n.d. American Vacuum Society.
- [6] BAETSL, L. and RAEDT, C. D., “Limitations of actinide recycle and fuel cycle consequences: a global analysis part 1: Global fuel cycle analysis,” *Nuclear Engineering and Design*, vol. 168, pp. 191–201, 1997.
- [7] BAETSL, L. and RAEDT, C. D., “Limitations of actinide recycle and fuel cycle consequences. a global analysis part 2: Recycle of actinides in thermal reactors: impact of high burn up lwr-uo 2 fuel irradiation and multiple recycle of lwr-mox fuel on the radiotoxic inventory,” *Nuclear Engineering and Design*, vol. 168, pp. 203–210, 1997.
- [8] BAETSLE, L. H., WAKABAYASHI, T., and SAKURAI, S., “Status and assessment report on actinide and fission product partitioning and transmutation,” tech. rep., Nuclear Energy Agency, 1998.
- [9] BATES, J., “A model for the science of decision,” *Philosophy of Science*, vol. 21, no. 4, pp. 326–339, 1954.
- [10] BENEDICT, M., PIGFORD, T. H., and LEVI, H. W., *Nuclear Chemical Engineering*. McGraw-Hill Book Company, second ed., 1981.
- [11] BENEDICT, R. W., “Argonne pyroprocessing proposal.” Presentation for AAA Technical Quarterly Review, July 9-10, 2002.
- [12] BROOKS, H., “The resolution of technically intensive public policy disputes,” *Science, Technology, and Human Values*, vol. 9, no. 1, pp. 39–50, 1984.
- [13] BRWM, “Research needs for high level waste stored at tanks and bins at u.s. department of energy sites,” tech. rep., National Research Council, Board on Radioactive Waste Management (BRWM), 2001.
- [14] BRWM and DELS, “End points for spent nuclear fuel and high-level radioactive waste in russia and the united states,” tech. rep., National Research Council, Board on Radioactive Waste Management (BRWM) and Earth And Life Studies (DELS), National Academies Press 2003.

- [15] BRYANT, C. F., CHARLTON, W. S., LANDSBERGER, S., and VANDEGRIFT, G. F., "Developing computer models for solvent extraction processes for optimizing flow-sheets for actinide transmutation and analyzing their technical benefits/liabilities and proliferation resistance," November 10-12 2001.
- [16] BUNN, M., FETTER, S., HOLDREN, J. P., and VAN DER ZWAAN, B., "The economics of reprocessing vs. direct disposal of spent nuclear fuel," *Nuclear Technology*, vol. 150, no. 3, June 2005.
- [17] BUTLER, J., MORRICE, D. J., and MULLARKEY, P. W., "A multiattribute utility theory approach to ranking and selection," *Management Science*, vol. 47, no. 6, pp. 800–817, 2001.
- [18] CEA, "Long-lived radionuclides partitioned at will." Spring Newsletter, 2002. Commissariat à l'énergie atomique.
- [19] CHANDLER, J., "Pyroprocessing: Way of the future or modern alchemy?." Unpublished, 2005.
- [20] CLIFFORD, A. A., ZHU, S., SMART, N. G., LIN, Y., WAI, C. M., YOSHIDA, Z., MEGURO, Y., and ISO, S., "Modelling the extraction of uranium with supercritical carbon dioxide," *Journal of Nuclear Science and Technology*, vol. 38, no. 6, pp. 433–438, 2001.
- [21] COLLINS, E. D., BENKER, D. E., SPENCER, B. B., BARON, P., DINH, B., BOND, W. D., and CAMPBELL, D. O., "Development of the urex+ co-decontamination solvent extraction process," November 16-20 2003.
- [22] COLLINS, E., "Closing the fuel cycle can extend the lifetime of the high-level-waste repository," in *American Nuclear Society 2005 Winter Meeting, Washington D.C.*, p. November 17, 2005.
- [23] CONGRESS, U., "Energy and water development appropriations for fy 2006," 2005. HR 2419.
- [24] CONGRESS, U., "Nuclear fuel reprocessing." U.S. House of Representatives Committee on Science Subcommittee on Energy, June 16 2005.
- [25] COWAN, R., "Nuclear power reactors: A study in technological lock-in," *Journal of Economic History*, vol. 50, no. 3, pp. 541–567, 1990.
- [26] CPSMA, "Electrometallurgical techniques for doe spent fuel treatment: Status report on argonne national laboratory's r and d activity as of fall 1998," tech. rep., National Research Council, Commission on Physical Sciences Mathematics and Applications, 1999.
- [27] CPSMA, "Electrometallurgical techniques for doe spent fuel treatment: Final report," tech. rep., National Research Council, Commission on Physical Sciences Mathematics and Applications, 2000.
- [28] DE REGGE, P. and BODEN, R., "Review of chemical separation techniques applicable to alpha spectrometric measurements," in *Nucl. Instrum. Methods Phys. Res.*

- A, Accel. Spectrom. Detect. Assoc. Equip. (Netherlands)*, vol. 223, (Harwell, UK), pp. 181–7, 1984.
- [29] DENNISS, I. S. and JEAPES, A. P., “Reprocessing irradiated fuel,” in *The Nuclear Fuel Cycle: From Ore to Waste* (WILSON, P., ed.), Oxford Science Publications, 1996.
 - [30] DOE, “Advanced fuel cycle initiative: The future path for advanced spent fuel treatment and transmutation research,” tech. rep., Department of Energy, Office of Nuclear Energy Science and Technology, 2003.
 - [31] DOE, “Legacy story, chapter 2. nuclear weapons production processes and history.” electronic book, n.d. Department of Energy.
 - [32] DOWNAR, T. J. and SESONSKE, A., “Light water reactor fuel cycle optimization: Theory versus practice,” *Advances in Nuclear Science and Technology*, vol. 20, no. 1988, pp. 71–126, 1988.
 - [33] EARLY, T., LEE, C., LEE, J. K., LUSCOMBE, C., SMITH, C., STAMP, L., and TSANG, M., “Synthesis in supercritical carbon dioxide.” University of Cambridge, n.d. <http://www-holmes.ch.cam.ac.uk/content/co2/co2projects.html>, accessed July 2006.
 - [34] ENOKIDA, Y., EL-FATAH, S. A., and WAI, C. M., “Ultrasound-enhanced dissolution of UO_2 in supercritical CO_2 containing a CO_2 -philic complexant of tri-*n*-butyl phosphate and nitric acid,” *Industrial Eng. Chem. Research*, vol. 41, pp. 2282–2286, 2002.
 - [35] ENOKIDA, Y., TOMIOKA, O., LEE, S.-C., RUSTENHOLTZ, A., and WAI, C. M., “Characterization of a tri-*n*-butyl phosphate-nitric acid complex: A CO_2 -soluble extractant for dissolution of uranium dioxide,” *Industrial Engineering Chem. Research*, vol. 42, pp. 5037–5041, 2003.
 - [36] FUKASAWA, T., UMEHARA, H., and MATSUDA, M., “Nuclear fuel-cycle technologies for a long-term stable supply of energy,” *Hitachi Review*, vol. 50, pp. 89–95, 2001.
 - [37] GOLDSTEIN, M. K., “Methodology for comparative evaluation of nuclear fuel reprocessing techniques for advanced low proliferation risk fuel cycles,” in *Alternative Energy Sources: An International Compendium* (VEZIROGLU, T. N., ed.), vol. 5, Hemisphere Publishing Company, 1978.
 - [38] GOTOVCHIKOV, V. T., MAKAROV, V. I., OREKHOV, V. T., RYBAKOV, A. G., and SEREDENKO, V. A., “Decladding of irradiated fuel elements,” *Atomic Energy*, vol. 99, no. 4, 2005.
 - [39] GRIFFIN, N. L., “A clean technology approach for nuclear fuel reprocessing,” *Journal of Chemical Technology Biotechnology*, vol. 68, pp. 361–366, 1997.
 - [40] GRIFFITHS, T. R., VOLKOVICH, V. A., YAKIMOV, S. M., MAYC, I., SHARRAD, C. A., and CHARNOCK, J. M., “Reprocessing spent nuclear fuel using molten carbonates and subsequent precipitation of rare earth fission products using phosphate,” *Journal of Alloys and Compounds*, vol. 418, pp. 116–121, 2006.

- [41] GROVER, S. B. and KUMAR, J., "A review of the current concepts of radiation measurement and its biological effects," *Ind Journal Radiolo Imag*, vol. 12, no. 1, pp. 21–32, 2002.
- [42] HAMLETT, P. W., *Understanding technological politics: a decision-making approach*. Prentice-Hall, 1991.
- [43] IKEDA, H., "Safety design of purex liquids system: The flash and fire points," *Journal of Nuclear Science and Technology*, vol. 41, no. 4, pp. 534–536, 2004.
- [44] IKEGAMI, T., "Core concept of a compound fuel cycle," *Progress in Nuclear Energy*, vol. 47, no. 1-4, pp. 231–238, 2005.
- [45] INNOUE, T., "Actinide recycling by pyro-process with metal fuel fbr for future nuclear fuel cycle systems," *Progress in Nuclear Energy*, vol. 40, no. 3/4, pp. 547–554, 2002.
- [46] JIMENEZ, A., RAIOS-INSUA, S., and MATEOS, A., "A decision support system for multiattribute utility evaluation based on imprecise assignments," *Decision Support Systems*, vol. 36, pp. 65–79, 2003.
- [47] JIMENEZ, A., ROS-INSUA, S., and MATEOS, A., "A generic multi-attribute analysis system," *Computers and Operations Research*, vol. 33, pp. 1081–1101, 2006.
- [48] JOHNSON, T. R., LEWIS, M. A., NEWMAN, A., and LAIDLER, J. J., "Treatment of high-level wastes from the ifr fuel cycle," in *Proceedings of the American Chemical society Meeting*, pp. August 23–28, 1992.
- [49] JORDAN, G. M. and ROBERTS, L. E. J., "Environmental aspects of the fast reactor cycle, and discussion," *Philosophic Transactions of the Royal Society of London*, vol. 331, no. 1619, pp. 395–408, 1990.
- [50] KAMOSHIDA, M., KAWAMURA, F., SASAHIRA, A., FUKASAWA, T., TSAWA, and YAMASHITA, J., "A new concept for the nuclear fuel recycle system: Application of the fluoride volatility reprocessing," *Progress in Nuclear Energy*, vol. 37, no. 1-4, pp. 145–150, 2000.
- [51] KANG, J. and VON HIPPEL, F., "Limited proliferation-resistance benefits from recycling unseparated transuranics and lanthanides from light-water reactor spent fuel," *Science and Global Security*, vol. 13, pp. 169–181, 2005.
- [52] KANTROWITZ, A., "Proposal for an institution for scientific judgement," *Science*, vol. 156, no. 3776, pp. 763–764, 1967.
- [53] KARRAKER, D., "Radiation chemistry of acetohydroxamic acid in the urex process," Tech. Rep. WSRC-TR-2002-00283, Westinghouse Savannah River Company, 2002.
- [54] KEENEY, R. L., "Assessment of multiattribute preferences," *Science*, vol. 168, no. 3938, pp. 1491–1492, 1970.
- [55] KEENEY, R. L., "Risk independence and multiattributed utility functions," *Econometrica*, vol. 41, no. 1, pp. 27–34, 1973.
- [56] KINOSHITA, K., "Development of pyro-process fuel cycle technology." CRIEPI News; Tokyo, Japan, 2002.

- [57] KO, W. I. and KIM, H. D., "Analysis of nuclear proliferation resistance of duple fuel cycle," *Journal of Nuclear Science and Technology*, vol. 38, no. 9, pp. 757–765, 2001.
- [58] KOBAYASHI, H., AMANO, O., KAWAMURA, F., AOI, M., HOSHINO, K., SASAHIRA, A., and KANI, Y., "Fluorex reprocessing system for the thermal reactors cycle and future thermal/fast reactors (coexistence) cycle," *Progress in Nuclear Energy*, vol. 47, no. 1-4, pp. 380–388, 2005.
- [59] KOYAMA, T., "Present status of the advanced aqueous separation process technology development." International Symposium NUCEF2005, February 9-10, presentation, 2005.
- [60] KULCINSKI, "Nuclear engineering materials course notes." University of Wisconsin, 1999. <http://fti.neep.wisc.edu/neep423/FALL99/lecture5.pdf>, accessed February 2006.
- [61] KUZNETSOV, S., HAYASHI, H., MINATO, K., and GAUNE-ESCARD, M., "Determination of uranium and rare-earth metals separation coefficients in UCl₃-KCl melt by electrochemical transient techniques," *Journal of Nuclear Materials*, vol. 344, pp. 169–172, 2005.
- [62] LAGUS, T., "Reprocessing of spent nuclear fuel: A policy analysis," tech. rep., University of Minnesota, ASME, August 4 2005.
- [63] LAIDLER, J. J., BATTLES, J. E., MILLER, W. E., ACKERMAN, J. P., and CARLS, E. L., "Development of pyroprocessing technology," *Progress in Nuclear Energy*, vol. 31, pp. 131–140, 1997.
- [64] LAIDLER, J. J., BURRIS, L., COLLINS, E., DUGUID, J., HENRY, R. N., HILL, J., KARELL, E., MCDEAVITT, S., THOMPSON, M., WILLIAMSON, M. A., and WILLIT, J. L., "Chemical partitioning technologies for an atw system," *Progress in Nuclear Energy*, vol. 38, no. 1/2, pp. 65–79, 2001.
- [65] LAPORTE, T. R., "Nuclear waste: Increasing scale and sociopolitical impacts," *Science*, vol. 201, no. 4350, pp. 22–28, 1978.
- [66] LAW, J. D., HERBST, R. S., TODD, T. A., ROMANOVSKIY, V. N., BABAIN, V. A., ESIMANTOVSKIY, V. M., SMIRNOV, I. V., and ZAITSEV, B. N., "The universal solvent extraction process ii: Flowsheet development and demonstration of the unex process for the separation of cesium, strontium, and actinides from actual acidic radioactive waste," *Solvent Extraction and Ion Exchange*, vol. 19, no. 1, pp. 23–36, 2001.
- [67] LAW, J., HERBST, S., PETERMAN, D., and TODD, T., "Development of a cobalt dicarbollide/polyethylene glycol solvent extraction process for separation of cesium and strontium in support of advanced aqueous reprocessing," *Nuclear Technology*, vol. 147, pp. 284–290, 2004.
- [68] LEVHARI, D., "Extensions of Arrow's "learning by doing"," *Review of Economic Studies*, vol. 33, no. 2, pp. 117–131, 1966.
- [69] LEVITZ, N. M., "A conceptual design study of a fluoride-volatility plant for reprocessing LMFBR fuels," tech. rep., Atomic Energy Commission, 1969.

- [70] MASSET, P., BOTTOMLEY, D., KONINGS, R., MALMBECK, R., RODRIGUES, A., SERP, J., and GLATZ, J.-P., “Electrochemistry of uranium in molten licl-kcl eutectic,” *Journal of the Electrochemical Society*, vol. 152, no. 6, pp. A1109–A1115, 2005.
- [71] MCPHEETERS, C., PIERCE, R., and MULCAHEY, T., “Application of the pyrochemical process to recycle of actinides from lwr spent fuel,” *Progress in Nuclear Energy*, vol. 21, no. 1/2, pp. 175–186, 1997.
- [72] MILLER, J. R., CLARK, R. G., MARSH, W. R., DEMINICO, R. J., and HOYT, R. C., “Airox dry pyrochemical processing of oxide fuels - proliferation resistant reprocessing method,” *Abstracts of Papers of the American Chemical Society*, no. APR, pp. 126–126, 1979.
- [73] MURALI, M. S. and MATHUR, J. N., “Use of a mixture of trpo and tbp for the partitioning of actinides from high-level waste solutions of purex origin and its comparison with cmpo and other phosphorus-based extractants,” *Solvent Extraction and Ion Exchange*, vol. 19, no. 1, pp. 61–77, 2001.
- [74] MURRAY, R. L. R., *Nuclear energy: an introduction to the concepts, systems, and applications of nuclear processes*. Butterworth-Heinemann, fifth ed., 2001.
- [75] NEA, “Actinide separation chemistry in nuclear waste streams and materials,” Tech. Rep. NEA/NSC/DOC(97)19, Nuclear Energy Agency, 1997.
- [76] NEA, “Partitioning and transmutation of minor actinides and fission products,” 2001.
- [77] NEA, “Trends in the nuclear fuel cycle,” tech. rep., Organisation for Economic Co-operation and Development (OECD) and Nuclear Energy Agency (NEA), 2002.
- [78] NEA, “Working group on flowsheet studies.” Nuclear Energy Agency, 2006. [<http://www.nea.fr/html/science/wpfc/wg-flowsheet.html>], accessed June 2006.
- [79] NEI, “Plutonium and uranium processing fact sheet.” Nuclear Energy Institute, 2003. <http://www.nei.org>, accessed January 2006.
- [80] NEVADA, “Molten salt process.” University of Nevada, 2002. Lecture.
- [81] NIST, “Nist standard reference data base 7.” National Institute for Standards and Technology, n.d.
- [82] NRC, “Fact sheet on dirty bombs.” Nuclear Regulatory Commission, 2003.
- [83] OCRWM, “Nuclear waste policy act, as amended,” tech. rep., Department of Energy, Office of Civilian Radioactive Waste Management (OCRWM), 2004.
- [84] OGAWA, T. and MINATO, K., “Dissolution and formation of nuclear materials in molten salts,” *Pure and Applied Chemistry*, vol. 73, no. 5, pp. 799–806, 2001.
- [85] OWCA, W. A. and MAJUMDAR, D., “Airox recycling for the us: to be or not to be?,” in *Proceedings of the International Conference and Technology Exposition on Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options. Global '93*, vol. vol.2, (Seattle, WA, USA), pp. 729–32, ANS, 1993.

- [86] PEREIRA, C., VANDEGRIFT, G., REGALBUTO, M., AASE, S., BAKEL, A., BOWERS, D., BYRNES, J., CLARK, M., EMORY, J., FALKENBERG, J. R., GELIS, A., HAFENRICHTER, L., LEONARD, R., QUIGLEY, K., TSAI, Y., POL, M. V., and LAIDLDER, J., "Lab-scale demonstration of the urex+2 process using spent fuel," in *Waste Management Conference, Tucson, AZ*, pp. February 27 – March 3, 2005.
- [87] PIERCE, R. A., THOMPSON, M. C., and RAY, R. J., "Solubility limits of dibutyl phosphoric acid in uranium-nitric acid solutions," *Separation Science and Technology*, vol. 36, no. 5-6, pp. 767–783, 2001.
- [88] RAJ, B. and MUDALI, U. K., "Materials development and corrosion problems in nuclear fuel reprocessing plants," *Progress in Nuclear Energy*, vol. 48, pp. 283–313, 2006.
- [89] REGALBUTO, M. C., "Chemical reprocessing plant simulation," December 14-16 2006.
- [90] REGALBUTO, M. C., COPPLE, J. M., LEONARD, R., PEREIRA, C., and VANDEGRIFT, G. F., "Solvent extraction process development for partitioning and transmutation of spent fuel," in *Actinide and Fission Product Partitioning and Transmutation Eighth Information Exchange Meeting, Las Vegas, NV*, pp. November 9 – 11, 2004.
- [91] RUDISILL, T. S., THOMPSON, M. C., NORATO, M. A., KESSINGER, G. F., PIERCE, R. A., and JOHNSON, J. D., "Demonstration of the urex solvent extraction process with dresden reactor fuel," Tech. Rep. WSRC-MS-2003-00089, Rev. 1, Westinghouse Savannah River Company, 2003.
- [92] RUYAT, Y., NAKAYAMA, Y., MATSUMURA, T., WAKABAYASHI, K., OKADA, K., MIYAKI, A., OGAWA, T., and YOSHIDA, M., "Explosion strength of tri-n-butyl phosphate and fuming nitric acid (tbp/fna) mixture evaluated by underwater explosion tests," *Science and Technology of Energetic Materials*, vol. 65, no. 1, pp. 14–20, 2004.
- [93] SAKAMURA, Y., KURATA, M., USAMI, T., and INOUE, T., "Conversion of oxide into metal or chloride for the pyrometallurgical partitioning process," 2004.
- [94] SCHMIEDER, H. and GALLA, U., "Electrochemical process for nuclear fuel reprocessing," *Journal of Applied Electrochemistry*, vol. 30, pp. 201–207, 2000.
- [95] SCHURHAMMER, R. and WIPFF, G., "Effect of the tbp and water on the complexation of uranyl nitrate and the dissolution of nitric acid into supercritical co₂. a theoretical study," *Journal of Physical Chemistry*, vol. 109, pp. 5208–5216, 2005.
- [96] SELVADURAY, G. S., *Comparative evaluation of nuclear fuel reprocessing techniques for advanced fuel cycle concepts*. PhD thesis, Stanford University, 1978.
- [97] SELVADURAY, G. S., GOLDSTEIN, M. K., and ANDERSON, R. N., "Separation technologies reviewed," *Nuclear Engineering International*, vol. 23, no. 274, pp. 35–40, 1978.
- [98] SERRANO-PURROY, D., BIRCK, S., CHRISTIANSEN, B., GLATZ, J.-P., HOLLAS, S., MALMBECK, R., RMER, K., SHEPLER, C., TEIXEIRA, E., WEERD, W. D., and WINCKEL, S. V., "Towards a diamex process flowsheet using high active concentrate (hac)," October 14-16 2002.

- [99] SERRANO-PURROY, D., BARON, P., CHRISTIANSEN, B., GLATZ, J.-P., MALMBECKA, C. M., and MODOLO, G., "First demonstration of a centrifugal solvent extraction process for minor actinides from a concentrated spent fuel solution," *Separation and Purification Technology*, vol. 45, pp. 157–162, 2005.
- [100] SHROPSHIRE, D. and CHANDLER, J., "Financing strategies for a nuclear fuel cycle facility," in *14th International Conference on Nuclear Engineering*, (Miami, Florida), 2006.
- [101] SPINRAD, B. I., "Plutonium recycling," *Science*, vol. 237, no. 4816, pp. 707–708, 1987.
- [102] STEVENS, C. E., *The EBR-II fuel cycle story*. American Nuclear Society, 1987.
- [103] SUN, L., LI, J., CHEN, Y., CLARKSEAN, R., PEPPER, D. W., LAIDLER, J., and VANDEGRIFT, G., "Development of a systems engineering model of the chemical separations process," June 9-13 2002.
- [104] TAHRAOUI, A. and MORRIS, J. H., "Decomposition of solvent extraction media during nuclear reprocessing: Literature review," *Separation Science and Technology*, vol. 30, no. 13, pp. 2603–2630, 1995.
- [105] TEDDER, D. D. W., *Preliminary Chemical Process Design and Economics*. Hickory Mountain, 2005. Georgia Institute of Technology.
- [106] THOMAS, T. R., "Airox nuclear fuel recycling and waste management," in *Proceedings of the International Conference and Technology Exposition on Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options. Global '93*, vol. vol.2, (Seattle, WA, USA), pp. 722–8, ANS, 1993.
- [107] TOMIOKA, O., MEGURO, Y., ISO, S., YOSHIDA, Z., ENOKIDA, Y., and YAMAMOTO, I., "New method for the removal of uranium from solid wastes with supercritical co₂ medium containing hno₃-tbp complex," *Journal of Nuclear Science and Technology*, vol. 38, no. 6, pp. 461–462, 2001.
- [108] ULSTRUP, J., "Methods of separating actinide elements," *International Atomic Energy Agency – Atomic Energy Review*, vol. 4, no. 4, pp. 3–82, 1966.
- [109] UOZUMI, K., "High throughput electrorefining of uranium in pyro-reprocessing," 2004.
- [110] UOZUMI, K., KINOSHITA, K., INOUE, T., FUSSELMAN, S., GRIMMETT, D., ROY, J., STORVICK, T., KRUEGER, C., and NABELEK, C., "Pyrometallurgical partitioning of uranium and transuranic elements from rare earth elements by electrorefining and reductive extraction," *Journal of Nuclear Science and Technology*, vol. 38, no. 1, pp. 36–44, 2001.
- [111] ZABUNOGLU, O. H. and OZDEMIR, L., "Purex co-processing of spent lwr fuels: flow sheet," *Annals of Nuclear Energy*, vol. 32, pp. 151–162, 2005.